

Title: Advanced Hydrogen Transport Membranes for Vision 21 Fossil Fuel Plants

Type of Report: Quarterly

Reporting Period Start Date: January 2, 2001

Reporting Period End Date: April 1, 2001

Principal Authors: Shane E. Roark, Tony F. Sammells, Adam E. Calihman, Lyrik Y. Pitzman, Pamela M. Van Calcar, Richard A. Mackay, Tom F. Barton, Sara L. Rolfe, Richard N. Kleiner, James E. Stephan, Tim R. Armstrong, Mike J. Holmes, Aaron L. Wagner

Date Report was Issued: April 30, 2001

DOE Award Number: DE-FC26-00NT40762

Name and Address of Submitting Organization:

Eltron Research Inc., 4600 Nautilus Court South, Boulder, CO 80301-3241

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

ABSTRACT

Eltron Research Inc., and team members CoorsTek, McDermott Technology, Inc., Süd Chemie, Argonne National Laboratory, and Oak Ridge National Laboratory are developing an environmentally benign, inexpensive, and efficient method for separating hydrogen from gas mixtures produced during industrial processes, such as coal gasification. This project was motivated by the National Energy Technology Laboratory (NETL) Vision 21 initiative which seeks to economically eliminate environmental concerns associated with the use of fossil fuels. This objective is being pursued using dense membranes based in part on Eltron-patented ceramic materials with a demonstrated ability for proton and electron conduction. The technical goals are being addressed by modifying single-phase and composite membrane composition and microstructure to maximize proton and electron conductivity without loss of material stability. Ultimately, these materials must enable hydrogen separation at practical rates under ambient and high-pressure conditions, without deactivation in the presence of feedstream components such as carbon dioxide, water, and sulfur.

During this quarter, it was demonstrated that increasing the transition metal loading in a model perovskite composition resulted in an increase in hydrogen flux. Improved flux corresponded to the emergence of additional phases in the ceramic membrane, and highest flux was achieved for a composite consisting of pseudo-cubic and rhombohedral perovskite phases. A 0.9-mm thick membrane of this material generated a hydrogen flux in excess of 0.1 mL/min/cm², which was approximately 35 times greater than analogs with lower transition metal levels. The dopant level and crystal structure also correlated with membrane density and coefficient of thermal expansion, but did not appear to affect grain size or shape. Additionally, preliminary ceramic-metal (cermet) composite membranes demonstrated a 10-fold increase in flux relative to analogous membranes composed of only the ceramic component. The hydrogen flux for these cermet samples corresponded to a conductivity of $\sim 10^{-3}$ S/cm, which was consistent with the predicted proton conductivity of the ceramic phase.

Increasing the sweep gas flow rate in test reactors was found to significantly increase hydrogen flux, as well as “apparent” material conductivity for all samples tested. Adding humidity to the feed gas stream produced a small increase in hydrogen flux. However, the catalyst on ceramic membrane surfaces did not affect flux, which suggested that the process was membrane-diffusion limited.

Representative samples and fabrication processes were evaluated on the basis of manufacturing practicality. It was determined that optimum membrane densification occurs over a very narrow temperature range for the subject ceramics. Additionally, calcination temperatures currently employed result in powders that are difficult mill and screen. These issues must be addressed to improve large-scale fabricability.

INTRODUCTION

The objective of this project is to develop an environmentally benign, inexpensive, and efficient method for separating hydrogen from gas mixtures produced during industrial processes, such as coal gasification. This objective will be accomplished by employing dense ceramic and composite membranes based in part on Eltron-patented materials (Patent No. US5821185) with a demonstrated ability for rapid proton and electron conduction. The primary technical challenge in achieving the goals of this project will be to optimize membrane composition and microstructure to enable practical hydrogen separation rates and chemical stability. Other key aspects of this developing technology include catalysis, ceramic processing methods, and separation unit design operating under high pressure. To achieve these technical goals, Eltron Research Inc. has organized a consortium consisting of CoorsTek, McDermott Technology, Inc. (MTI), Süd Chemie, Inc. (SCI), Argonne National Laboratory (ANL), and Oak Ridge National Laboratory (ORNL).

During this quarter, it was demonstrated that increasing the transition metal loading in a model perovskite composition resulted in an increase in hydrogen flux. Improved flux corresponded to the emergence of additional phases in the ceramic membrane, and highest flux was achieved for a composite consisting of pseudo-cubic and rhombohedral perovskite phases. Additionally, preliminary ceramic-metal (cermet) composite membranes demonstrated a 10-fold increase in flux relative to analogous membranes composed of only the ceramic component. The hydrogen flux for these cermet samples corresponded to a conductivity of $\sim 10^{-3}$ S/cm, which was consistent with the predicted proton conductivity of the ceramic phase.

The effect of experimental variable on hydrogen flux also was investigated. Increasing the sweep gas flow rate in test reactors was found to significantly increase hydrogen flux, as well as “apparent” material conductivity for all samples tested. Adding humidity to the feed gas stream produced a small increase in hydrogen flux. However, the catalyst on ceramic membrane surfaces did not affect flux, which suggested that the process was membrane-diffusion limited.

Representative samples and fabrication processes were evaluated on the basis of manufacturing practicality. It was determined that optimum membrane densification occurs over a very narrow temperature range for the subject ceramics. Additionally, calcination temperatures currently employed result in powders that are difficult to mill and screen. These issues must be addressed to improve large-scale fabricability.

Other issues discussed in this report include stability and mechanical testing, advanced separation unit design, commercial concepting, high-pressure seals, and thin-film membranes.

EXPERIMENTAL

The Experimental Section in the first quarterly report (January 1, 2001) contained detailed descriptions of equipment and procedures to be used over the duration of this program. The specific aspects presented were: (a) preparation of ceramic powders, (b) preparation of composite materials, (c) fabrication of tube and disk membranes, (d) construction and operation of conductivity apparatus, (e) construction and operation of ambient-pressure hydrogen separation units, (f) construction and operation of high-pressure hydrogen separation units, (g) hydrogen transport and ambipolar conductivity measurements and calculations, and (h) fabrication of thin film ceramics. For brevity, these general issues will not be repeated. However, modification of equipment or methods, as well

as other experimentally relevant issues, will be reported in the Results and Discussion section under their corresponding Tasks as outlined in the original proposal.

RESULTS AND DISCUSSION

Tasks 1 & 2 *Preparation, Characterization, and Evaluation of Hydrogen Transport Membranes*

Contributors: Eltron, CoorsTek, ORNL, MTI, SCI

a. **Effect of Dopant Concentration on Model Perovskite Hydrogen Transport Membranes - Eltron**

A series of relatively thick tube membranes (~ 0.9 mm) were tested with the general composition $AB_{1-x}B_x'O_{3-\delta}$, where B' represents a transition metal. Figure 1 shows hydrogen transport versus temperature for the composition $AB_{0.7}B_{0.3}'O_{3-\delta}$, which demonstrated the best performance of the series. The transport/temperature curves were characterized by an increase in hydrogen flux between ~ 600° and 850°C, followed by a decrease at temperatures greater than ~ 900°C. Figure 2 compares the maximum transport rate as a function of transition metal dopant concentration (x in $AB_{1-x}B_x'O_{3-\delta}$) for a series of tube membranes tested under identical conditions. From x = 0.05 to 0.2,

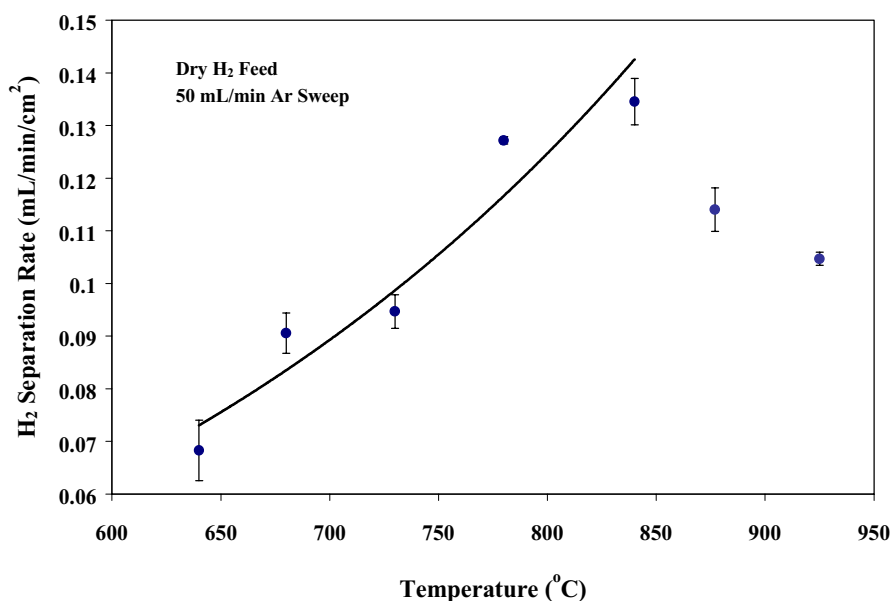


Figure 1. Plot of hydrogen transport rate versus temperature for a tube membrane with the empirical composition $AB_{0.7}B_{0.3}'O_{3-\delta}$. The membrane was 0.9 mm thick. The flow rate was 50 mL/min for both the hydrogen feed and argon sweep gases. The catalyst was porous platinum.

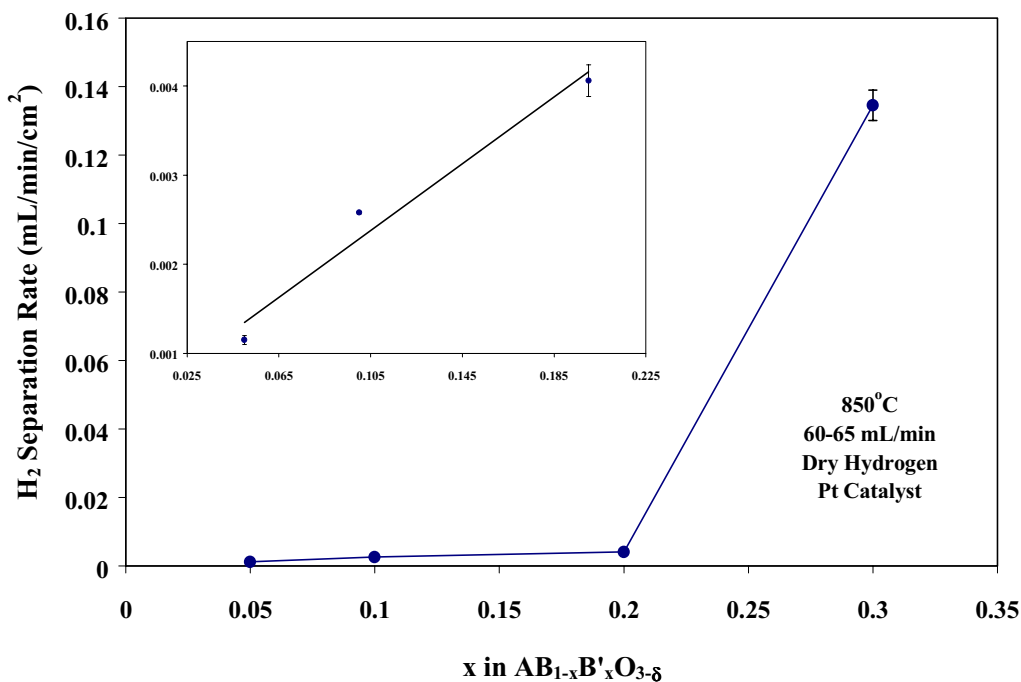


Figure 2. Relationship between maximum hydrogen transport and dopant concentration for tube membranes with the general formula $AB_{1-x}B'_xO_{3-\delta}$. The inset in the figure shows an expanded plot of the first three points. Each point was taken at 850°C using dry hydrogen as the feed and argon as the sweep gas. The flow rate for both gases was 60 - 65 mL/min. The catalyst was porous platinum.

there was approximately a four-fold increase in transport. For clarity, those three points are shown expanded in the figure inset. However, increasing x to 0.3 generated approximately a 35-fold increase in transport. X-ray diffraction patterns indicated significant phase differences between these samples. Specifically, at $x = 0.05$ the material was primarily a single-phase pseudo-cubic perovskite with a lattice parameter of 4.393(1) Å. This value was approximately equal to the undoped ABO_3 analog. At $x = 0.1$, a second unidentified phase was evident, which became more prominent at $x = 0.2$. However, at $x = 0.3$ the unidentified second phase was completely absent and a new phase appeared that was consistent with a rhombohedral perovskite with lattice parameters $a = 5.797$ Å and $c = 28.595$ Å. Each composition had essentially the same lattice parameter for the pseudo-cubic perovskite phase ($a = 4.39$ Å), indicating minimal substitution of the transition metal into the cubic perovskite lattice.

Density measurements were obtained for three to twelve samples of each composition using the Archimedes' method, and the results are shown in Figure 3. The undoped perovskite (not shown) had a density of 5.87 ± 0.03 g/cm³; thus, addition of a transition metal dopant significantly increased sample density. However, the greatest increase in density was attained for the sample with $x = 0.05$, and further doping decreased density concurrent with the emergence of the second

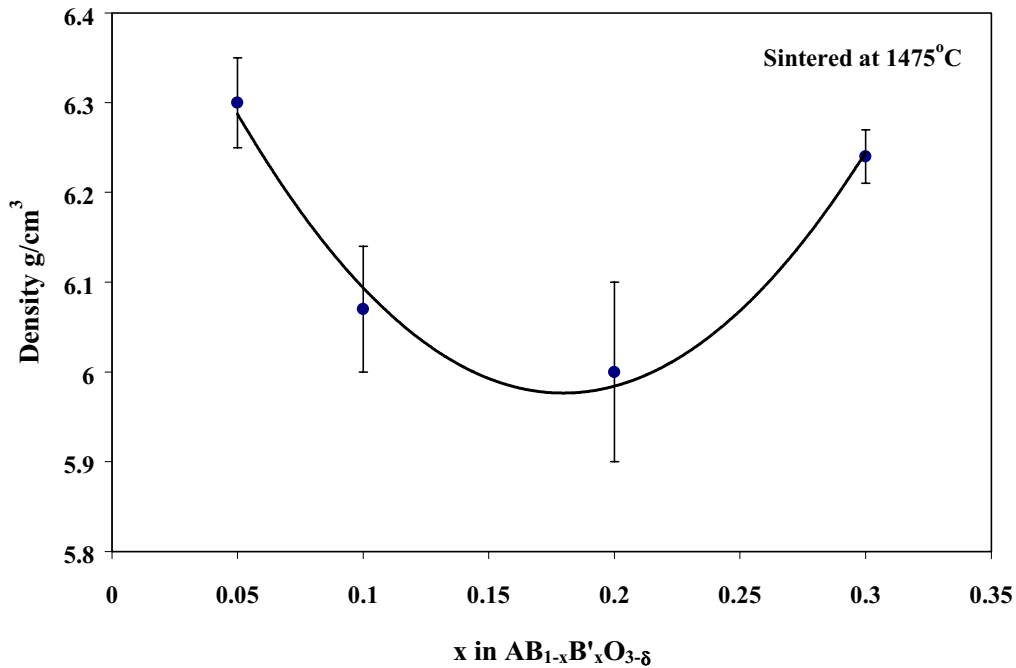


Figure 3. Plot showing the effect of transition metal doping level on sample density. The error bars represent $\pm 1\sigma$ from 3 to 12 measurements using the Archimedes method. All samples were sintered at 1475°C for 4 hrs.

crystallographic phase. At the highest transition metal level ($x = 0.3$) there was an increase in density and, as indicated above, an additional phase was present in the XRD pattern.

A representative scanning electron microscope (SEM) image is shown in Figure 4, and the average grain diameter for each sample is plotted as function of doping level in Figure 5. The grain sizes were roughly between 20 and 40 μm in diameter, and as evident in the plot, there was no apparent relationship between grain size and doping level. Also, there were no obvious qualitative differences in the grain shapes.

The coefficient of thermal expansion (CTE) was determined for each sample using sections of tube membranes between 6 and 18 mm in length. Heating and cooling cycles were performed between 100° and 950°C at 5°C/min under nitrogen. Below about 600°C, there was considerable hysteresis in the curves for heating and cooling. However, at higher temperatures the CTE measurement were fairly stable. Figure 6 summarizes the expansion characteristics at 800°C as a function of dopant level. The values represent the averages of one heating and two cooling cycles. Although the CTE values between $x = 0.5$ and 0.2 were the same within error, the CTE was significantly lower at $x = 0.3$.

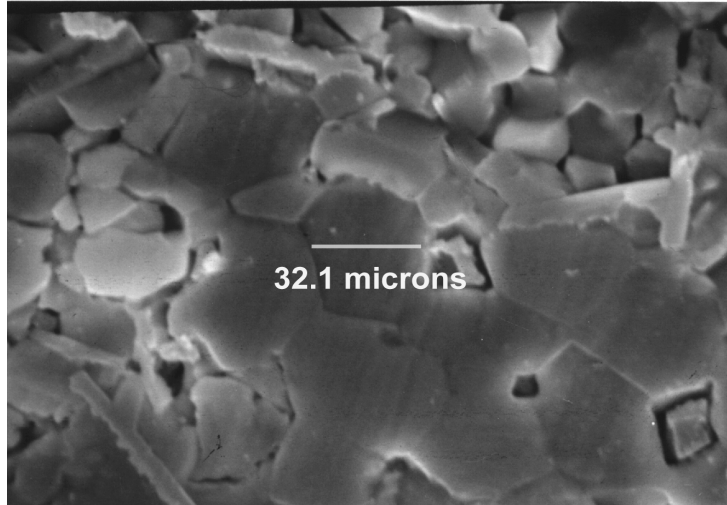


Figure 4. Representative SEM image showing grain sizes for a representative dense ceramic hydrogen transport membrane.

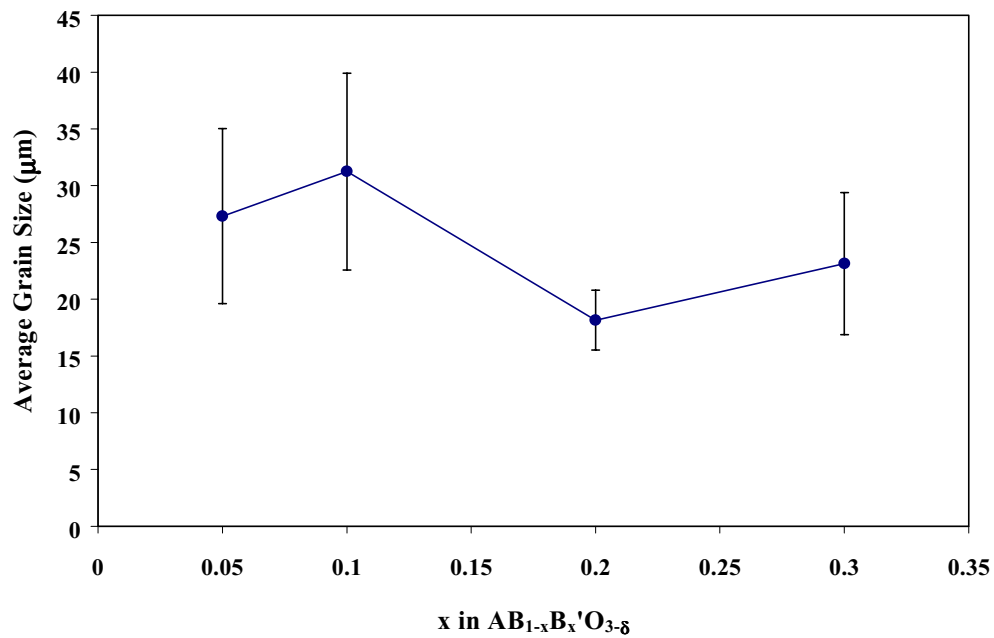


Figure 5. Plot showing grain diameter as a function of doping level for dense ceramic hydrogen transport membranes. The grain sizes are averages from 10 to 15 grain measurements for each sample, and the error bars ($\pm 1\sigma$) represent the range in grain sizes.

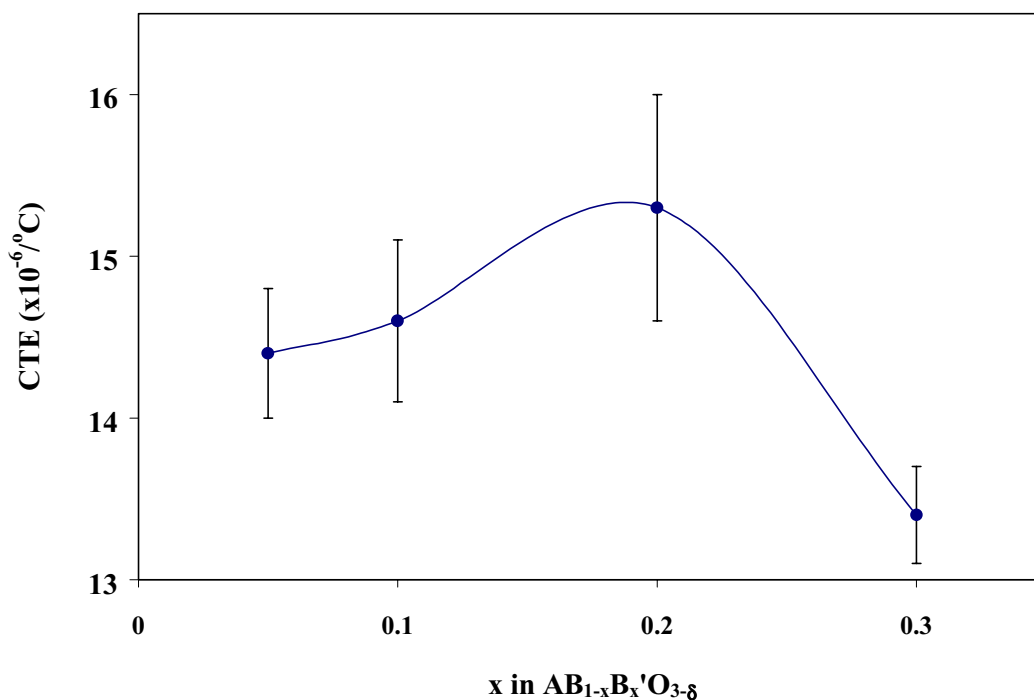


Figure 6. Plot showing the coefficient of thermal expansion for model perovskite hydrogen transport membranes as a function of dopant level. The values represent averages from one heating cycle and two cooling cycles. The error bars represent $\pm 1\sigma$.

b. Effect of Experimental Variables on Hydrogen Transport Through Dense Ceramic Membranes - Eltron

In an effort to identify the most appropriate experimental conditions for evaluating dense ceramic hydrogen transport membranes, hydrogen flux was measured as function of gas flow rate and moisture content. Figure 7 demonstrates that as the gas flow rate (on both sides of the membrane) was increased from 30 to 120 mL/min, there was a steady increase in hydrogen separation rate at each temperature tested. Since presumably increasing the sweep rate decreases hydrogen partial pressure, it is reasonable to expect a corresponding increase in potential and hydrogen flux. This presumption was supported by the 850°C data in Figure 8 A and B. However, at 800°C there was no significant difference in hydrogen concentration or potential between 65 and 120 mL/min, and at 750°C the hydrogen concentration actually increased slightly at 120 mL/min. When this data was used to calculate ambipolar conductivity (σ_{amb}), an apparent increase in conductivity with increasing flow rate was observed at each temperature, as shown in Figure 9. This dependence of apparent σ_{amb} on flow rate is typical, and underscores an experimental limitation when using bulk hydrogen partial pressure to represent the actual interfacial hydrogen partial pressure. It is possible that at lower flow rates there is a significant hydrogen partial pressure gradient between the membrane/gas phase interface and the bulk. Under these conditions, the

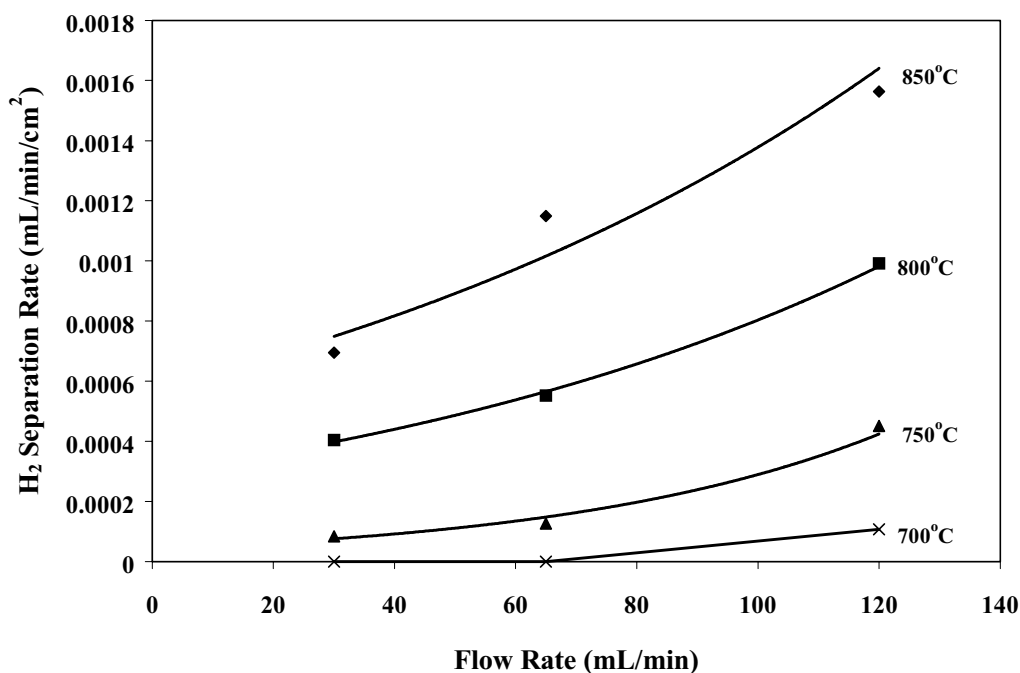


Figure 7. Plot showing the dependence of gas flow rate on hydrogen separation rate for several temperatures. Gases on both sides of the membrane were adjusted simultaneously to the values indicated.

apparent σ_{amb} calculated from the bulk hydrogen partial pressure will be lower than the actual σ_{amb} . As the sweep flow rate is increased and more permeate hydrogen is removed from the surface, the difference between hydrogen partial pressure in the bulk and interface likely is reduced, and the apparent σ_{amb} is closer to the actual value. Since the bulk hydrogen partial pressure values are not necessarily representative of the membrane interface, it is not unreasonable that hydrogen transport increased with sweep flow rate at 800° and 750°C although the calculated potentials did not show the expected trend.

Figure 10 shows a comparison of hydrogen transport rates using dry and humidified hydrogen. Humidified hydrogen (~2 to 5%) was prepared by bubbling the gas stream through water at room temperature prior to entering the separation cell. Approximately 5 hours were required after introducing humidity to achieve stable transport rates. Higher transport rates were observed with the humidified stream, and the improvement increased with temperature. However, the difference between the humid and dry stream was very small. Similar results were obtained for other membrane compositions, however, the effect of humidity on membranes with higher overall transport has not yet been tested. The improved separation rates in the presence of moisture presumably is due to preferential uptake of hydrogen through reaction of water with oxygen vacancies, relative to dry hydrogen.

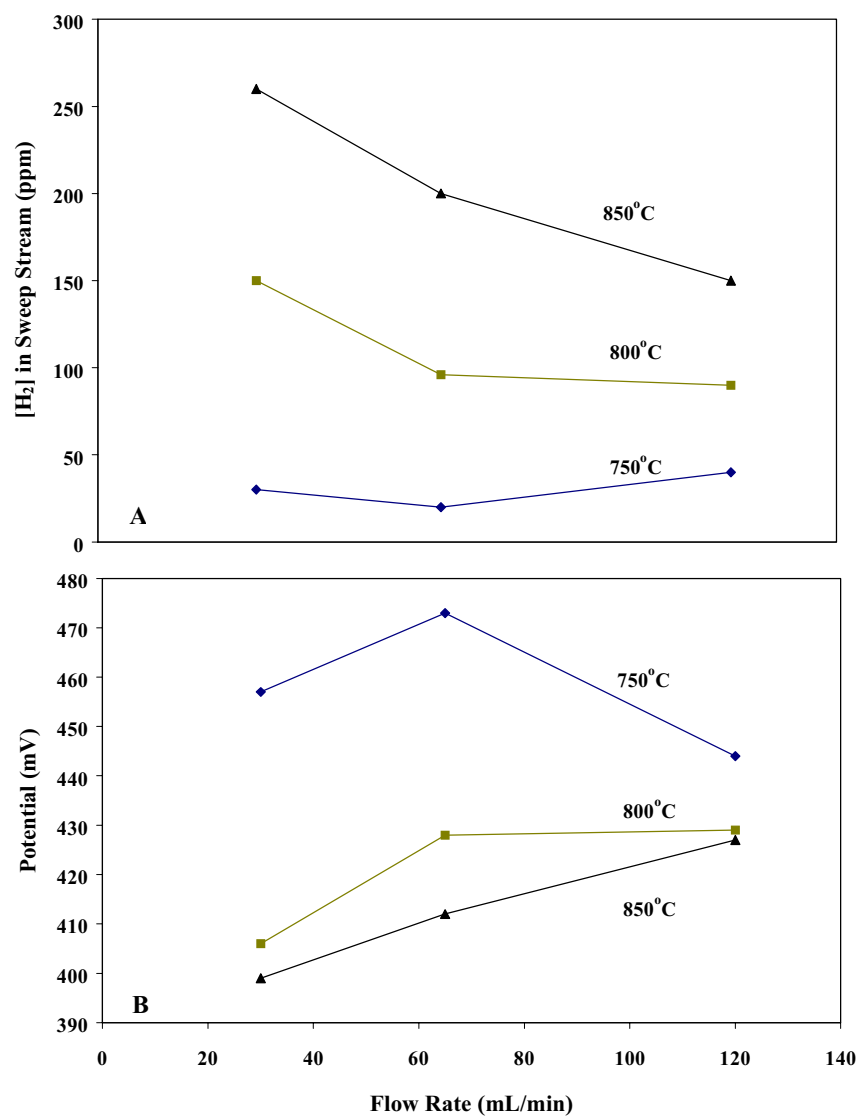


Figure 8. Plot (A) shows the concentration of hydrogen in the argon sweep stream as a function of sweep flow rate for several temperatures. The corresponding potentials across the membrane as calculated from the Nernst equation are shown in (B).

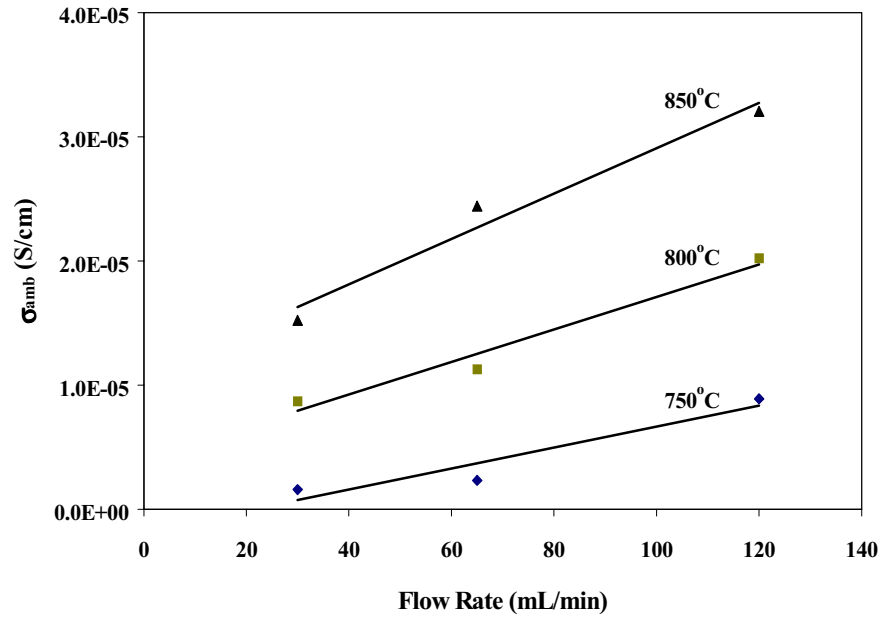


Figure 9. Plot showing the effect of sweep gas flow rate on calculated conductivity based on cell temperature and partial pressure of hydrogen in the exit stream.

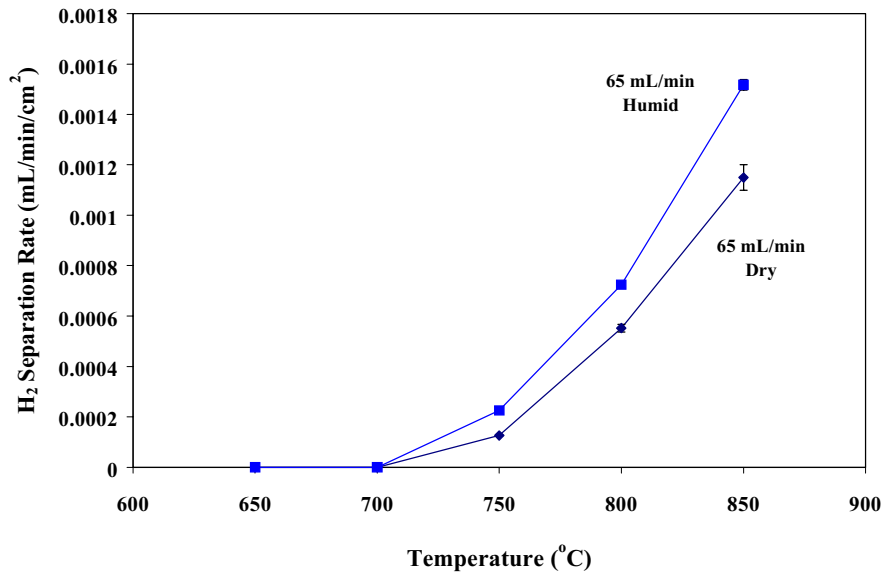


Figure 10. Plot comparing hydrogen transport rates in dry and humid hydrogen. Humidified gas was prepared by bubbling hydrogen through water at room temperature prior to entering the separation cell. The flow rate of hydrogen and sweep gas was 65 mL/min. The error bars represent $\pm 1\sigma$.

c. Effect of Catalyst on Hydrogen Transport Rates - Eltron, SCI

The majority of membranes tested so far have been thick enough (~1 mm) that hydrogen flux likely was limited by transport through the membrane, rather than by surface kinetics. Under this condition, it is not expected that the catalyst will have a dramatic effect on transport rates provided catalyst activity is above a certain level. Support for this presumption is shown in Figure 11. Both membrane samples in the figure were $AB_{0.9}B_{0.1}'O_{3-\delta}$, which was describe above in section (a). The Pt catalyst was a continuous highly porous film applied to the membrane surface, as described in the previous report. The 5 wt.-% Ni catalyst was prepared by impregnating ceramic powder (same composition as the membrane) with nickel nitrate dissolved in acetone. After drying and reductive calcination, the catalyst powder was mixed in toluene/ethanol with a binder and a plasticizer, then the slurry was painted onto the membrane. Within experimental variance, both catalysts performed about the same, although Ni was slightly better at lower temperatures and Pt was slightly better at high temperatures.

As indicated above, the membrane samples used for this experiment had very low transport, and more detailed catalyst experiments must be performed on thinner membranes of materials with higher conductivity. During the next quarter, these experiments will proceed at Eltron as well as SCI. Eltron will repeat the above experiments on other membrane compositions and SCI will focus on proprietary wet impregnation methods to increase catalyst metal dispersion. Specifically, SCI will deposit Ni as well as precious metals including Pt, Ru, Rh, Pd, Ir, and Os onto membrane samples prepared by Eltron. Eltron will test the catalyst-coated membranes for hydrogen transport

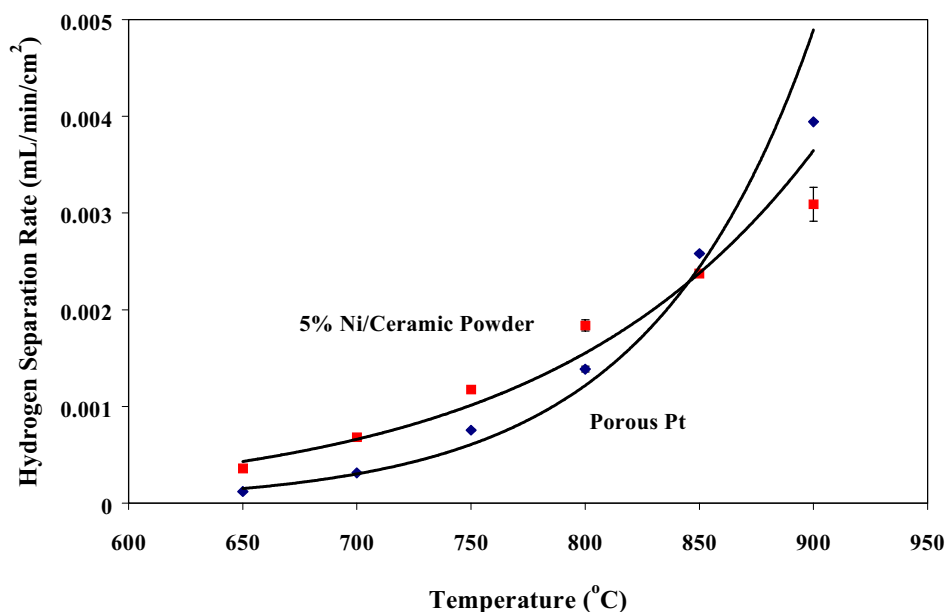


Figure 11. Plot showing hydrogen transport rate versus temperature for two $AB_{0.9}B_{0.1}'O_{3-\delta}$ membranes with different catalysts. The Pt catalyst was a continuous highly porous film applied to the membrane surface. The Ni catalyst was 5 wt.-% on a support powder applied to the membrane surface. Inlet and sweep gas flow rates were 60 mL/min.

as above.

d. Ceramic-Metal (CERMET) Composite Membranes - Eltron

Dense cermet membranes were prepared using a doped perovskite with the general composition $AB_{1-x}B_x'O_{3-\delta}$ as the ceramic phase. Numerous samples were prepared with varying ceramic/metal ratios and particle sizes. Large ceramic particles (0.4 - 1.5 μm) were prepared using solid-state synthesis without attrition, and small ceramic particles (40 nm) were prepared using coprecipitation. Large metal particles (3 - 7 μm) were purchased commercially either as the reduced metal or metal oxide. Small metal particles (50 nm) were achieved by precipitating the metal onto the ceramic powder from a metal nitrate precursor solution. Disk membranes prepared with 41, 44, and 47 wt.% metal all showed continuous metal phases in the electron micrographs, which was confirmed by testing electrical continuity using a voltmeter. As expected, smaller particle sizes led to smaller grain sizes in the densified membrane; however, there was no apparent difference in phase continuity using smaller particles relative to larger particles.

Figure 12 demonstrates the improvement in hydrogen flux upon compositing a mixed-conducting ceramic with a metal. The top curve is hydrogen flux across a 2.3-mm thick cermet membrane with 47 wt.% metal component and 53 wt.% mixed proton/electron conducting ceramic component. The bottom curve corresponds to a 1-mm thick membrane of the ceramic material only.

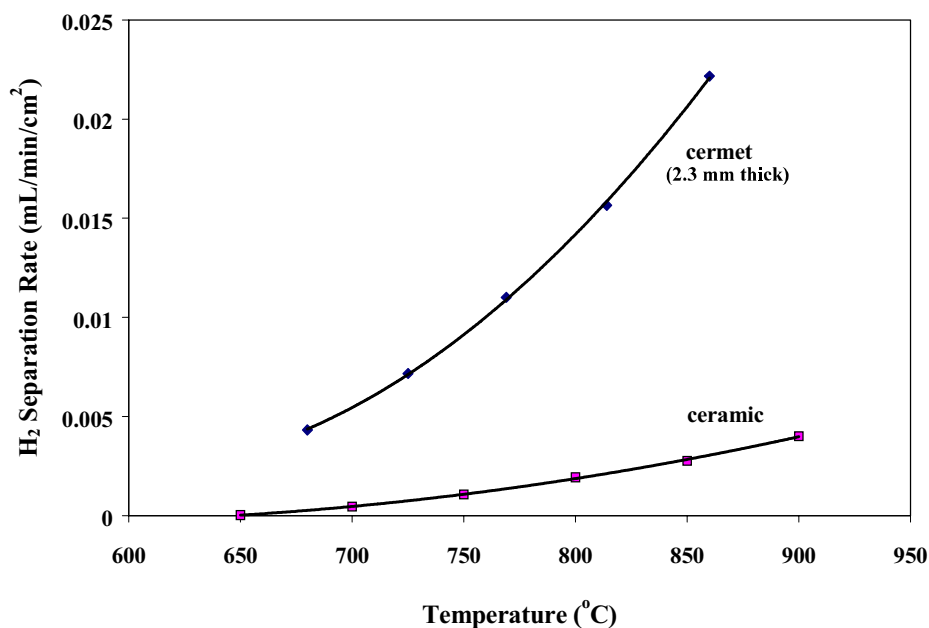


Figure 12. Plot showing the improvement in hydrogen flux upon compositing metal with a hydrogen-permeable ceramic. The cermet membrane (2.3 mm thick) was 47 wt.% metal, and the ceramic component was the exact same composition as the ceramic membrane (1 mm thick). The sweep gas (Ar) flow rate was 200 mL/min for the cermet and 230 mL/min for the ceramic.

The maximum hydrogen transport for the cermet membrane was one order of magnitude higher than the ceramic membrane. Similar improvement in hydrogen flux for cermet materials have recently been published by Balachandran *et al.* (Balachandran *et al.*; 26th Int. Tech. Conf. Coal Utilization and Fuel Sys., Clearwater, FL, March 5-8, 2001, pp's 751-761) Furthermore, it is significant that the cermet was more than twice as thick as the ceramic membrane. Using these hydrogen transport rates and membrane thicknesses, the apparent σ_{amb} for the cermet and ceramic was $\sim 10^{-3}$ S/cm and $\sim 10^{-4}$ S/cm, respectively. Since the cermet was not limited by electron conductivity, $\sigma_{amb} \approx \sigma_{H^+}$, which was consistent with ionic conductivity measurements obtained on the ceramic.

e. Manufacturing Issues and Relative Economics - CoorsTek

Part of Task 1 of this project involves outlining manufacturing issues and estimating the relative economics associated with large-scale production of the proposed ceramics or cermets. These goals are being pursued by CoorsTek, and involve i) reproducing Eltron's model materials and confirming equal performance, ii) reviewing membrane fabrication processes, iii) evaluating alternative processing methods, iv) assessing environmental aspects of processing methods, and v) investigating commercial sources and grades of starting materials to reduce production costs.

During this reporting period, a model perovskite compound was identified for initial manufacturing evaluation, and sample membranes were prepared at Eltron and CoorsTek to confirm reproducibility. This process involved uniaxial pressing and sintering of test parts at CoorsTek from powders supplied by Eltron, as well as from powders produced from raw materials acquired by CoorsTek. There was good agreement between the two test sets, however, parts produced from powders provided by Eltron had a slightly higher shrinkage rate (18.3 to 19.3 %) and density (94% of theoretical) than those prepared from CoorsTek powders (14.6 to 16.1% shrinkage, 90% of theoretical density). Studies are currently underway at CoorsTek to optimize densification.

Materials characterization also was performed in collaboration between Eltron and CoorsTek. Specifically, particle size measurements were performed on different instruments at the two facilities to identify target ranges for milling procedures. Additionally, XRD patterns were obtained on sintered parts to confirm equivalent crystal structures, and density measurements were performed by CoorsTek using three different techniques.

Two primary conclusions regarding manufacturing of model perovskite structures were derived from the above studies. First, the calcination temperatures used by Eltron result in powders that are very difficult to mill and screen. Second, the sintering temperature range to achieve maximum densification is very narrow. Accordingly, actions during the next quarter will include optimizing the calcination temperature and broadening the sintering range to increase the practicality of large-scale processing.

f. Stability and Mechanical Testing - ORNL

During this past quarter ORNL evaluated the stability of BaCeO₃ with Y and Zr additions in CO₂ to determine verify their phase stability. Powdered samples prepared by combustion synthesis were analyzed in a high-temperature x-ray diffractometer under flowing CO₂ to determine the temperature at which BaCO₃ forms. In all the samples containing Y, BaCO₃ formed at 1050°C. In samples containing >15mole % ZrO₂, no traces of BaCO₃ were detected.

The purpose of this study was to determine which compositions would be stable for long periods of time in air in order to enable analysis of their mechanical properties. Based on this work, 1 kg each of $\text{Ba}(\text{Ce}_{0.8}\text{Y}_{0.2})\text{O}_3$ and $\text{Ba}(\text{Ce}_{0.8}\text{Zr}_{0.2})\text{O}_3$ were purchased from Praxair Specialty Ceramics. These powders will be analyzed to determine: phase content, sinterability, and high-temperature mechanical properties in air including, fracture toughness, strength, elastic properties.

In addition, researchers at ORNL have been developing a method to test the materials under actual operating conditions and environments. ORNL has developed a biaxial flexure test in which the sample is sealed and one side exposed to H_2 and the other side to a higher oxygen partial pressure. The test cell is designed such that fracture can be initiated from either side to determine the impact chemical coefficient of expansion (oxygen nonstoichiometry) and defect distribution play in sample fracture.

g. Advanced Separation Device Design - ORNL

During this quarter ORNL completed development of the mathematical equations and codes to model constrained sintering of asymmetric membranes consisting of either: 1) a 2-layer membrane with a thin dense film supported on a thick porous substrate or 2) a 3-layer membrane with a thin dense film sandwiched between a thick porous substrate and a porous catalytic layer. The model was tested using existing data on a typical fuel cell system (yttria stabilized zirconia and Ni-zirconia cermet) and found to give very good results. The data required to test the model on BaCeO_3 will require 2 quarters to collect. Researchers at ORNL plan to initially model a 2-layer system. In this system the sintering rate of the porous layer varied such that in one case the rate will be controlled by additions of a fugative phase and in a second by coarsening the BaCeO_3 . In both cases, the thin membrane will be between 10 and 20 microns thick and sinter to full density at 1400°C (2 hrs). Its sintering properties will not be altered. This model will allow the team to determine the stresses that exist in the membrane after sintering as a function of the thickness of the layers, their elastic properties, and sintering rates.

h. Commercial Concepting - MTI

Work began on gathering background information to help define process conditions and to provide a starting point for assessing the economics for addition of a hydrogen separation system to a Vision 21 Plant. In addition to reviewing literature, a call was made to a contact from Kellogg, Brown & Root who has been working on site at the Power Systems Development Facility (PSDF) in Wilsonville, Alabama. The information from this contact and review of literature is being summarized by MTI in a memo to Eltron Research Inc. The topics include typical temperatures and pressures that can be expected for the separation membrane, the expected gas compositions, and trace species that the membrane will likely be exposed to. The memo will begin to address these issues as inputs to the membrane testing. Results from the membrane testing will then be used to help identify the acceptable range of process conditions for the reactor, so that viable process layouts can be confirmed.

Task 3 *High Pressure Hydrogen Separation*

Contributors: Eltron

Operation of dense ceramic hydrogen separation membranes at high temperatures and pressures requires a chemically resistant seal with similar mechanical and expansion characteristics as the membrane material. During this quarter, two seal compositions (designated as EL-1 and EL-2) were tested in four combinations. Seal formation was achieved by ramping the cell assembly up to 1000°C and holding for a period of time. The assembly then was cooled to 800°C for seal testing. Initially, nitrogen was used as the test gas, which only enabled a pressure of 15 psig to be maintained. However, by switching to a more reductive environment, a pressure of 47 psig was achieved without changing the seal composition.

Task 4 *Thin-Film Hydrogen Separation Membranes*

Contributors: Eltron

Thin films of model hydrogen transport ceramic membranes were prepared over a thickness range of 10 to 100 μm . Although some success in fabrication of these membranes was achieved during this quarter, there are still several processing issues that need to be addressed before these samples can be tested for hydrogen transport. To date, it has not been possible to achieve an adequate seal in the test reactor with thin film membranes. Furthermore, methods developed at Eltron for thin film oxygen separation ceramics must be modified to accommodate the different sintering characteristics of hydrogen transport ceramics.

Task 5 *Construction and Evaluation of Prototype Hydrogen Separation Unit*

No actions were performed on this task during this reporting period.

Task 6 *Membrane-Promoted Conversion of Alkanes to Olefins*

Contributors: Eltron

The laboratory-scale reactors have been constructed and this task currently is on schedule. However, actions associated with catalyst development and performance evaluation likely will be moved out to begin on month 12, rather than month 6 as indicated in the program timeline. This adjustment will enable Eltron and collaborators to take greater advantage of accomplishments in Tasks 1, 2, and 4.

OBJECTIVES FOR NEXT REPORTING PERIOD

Specific objectives for the next quarter are summarized as follows:

- Increase hydrogen permeation through cermet membrane by one order of magnitude.
- Reproduce and increase hydrogen permeation through composite ceramic membranes.
- Test a range of catalyst systems for promoting hydrogen transport. Determine if surface kinetics limit hydrogen flux.
- Demonstrate function of thin-film hydrogen separation membranes with acceptably low leak rates.
- Increase pressures maintainable by high-pressure seals by a factor of two.
- Continue with stability, design, manufacturing, and commercial concepting issues.
- Initiate neutron diffraction studies on model proton-conducting ceramics.

TIME LINES

The time lines separated into each task are presented below, with markers indicating overall progress for each subtask.

