CERAMIC MEMBRANE ENABLING TECHNOLOGY FOR IMPROVED IGCC EFFICIENCY

QUARTERLY TECHNICAL PROGRESS REPORT

Reporting Period Start Date: October 2000
Reporting Period End Date: December 2000

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Date Report was Issued: January 2001

DOE AWARD NO. DE-FC26-99FT40437

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ABSTRACT:

This quarterly technical progress report will summarize work accomplished for Phase 1 Program during the quarter October to December 2000. In task 1 careful modification of the processing conditions of the OTM has improved the properties of the final element. In addition, finite element modeling has been used to predict the mechanical behavior of OTM tubes and to identify strategies for improving OTM robustness. In task 2, composite elements of PSO1d have been prepared and tested for over 800 hours without degradation in oxygen flux. Alternative materials for composite OTM and architectures have been examined with success. In task 3, modification of fabrication routes has resulted in a substantial increase in the yield of PSO1d composite elements. The work in task 4 has demonstrated that composite OTM elements can produce oxygen at atmospheric pressure of greater than 95% purity from a high-pressure air feed gas. The work in task 5 to construct a multi-tube OTM reactor has begun.

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A. Executive Summary

The objectives of the second year of the program are to define a material composition and composite architecture that enable the oxygen flux and stability targets to be obtained in high-pressure flux tests. Composite development technology will be developed to enable the production of high-quality, dense membranes of a thickness that allows the oxygen flux target to be obtained. The fabrication technology will be scaled up to produce three feet composite tubes with the desired leak rate. A laboratory scale, multi-tube pilot reactor will be designed and constructed to produce oxygen.

In the first quarter of the second year of the program, work has focussed on materials optimization, composite and manufacturing development and oxygen flux testing at high pressures. This work has led to several major achievements, summarized below.

- **Oxygen has been produced from a composite OTM tube at the target purity level of greater than 95%.**
- Finite element analysis has predicted the mechanical behavior of OTM tubes and has suggested several strategies for increasing OTM robustness.
- Thin films of PSO1 have been successfully deposited on porous substrates of more commercially viable materials

B. Experimental Methods

B.1. OTM Materials Development Experimental Methods

Characterization of OTM and substrate materials has been undertaken using many different experimental procedures. These include permeation, crystallographic, thermomechanical, thermochemical and electrochemical measurements. Standard equipment such as XRD, SEM, dilatometry and TGA/DSC were used. In addition oxygen permeation testers were used to measure the oxygen flux of OTM discs. The permeation test facility was described in the DOE IGCC first annual report [1].

B.2. Composite OTM Development Experimental Methods

Various fabrication routes have been developed to prepare composite OTM samples. Small samples are first prepared and the fabrication routes that are most promising are further refined to enable larger OTM elements to be prepared. The fabrication routes used are proprietary information and included in the Appendix.

B.3. Manufacturing Development Experimental Methods

Fabrication routes developed in task 2 have been used for the manufacture of OTM elements for testing in the high-pressure permeation testers used in task 4.

B.4. Process Development Experimental Methods

Composite OTM elements of the required geometry prepared using methods developed in prior work have been tested for high temperature permeation utilizing the high-pressure test facility and method previously described in the DOE IGCC first annual report [1].

B.5. O-1 Pilot Reactor Development Experimental Methods

Commercial software has been used to determine mechanical stress and heat balance of the pilot scale reactor.

C. Results and Discussion

C.1. OTM Materials Development Results and Discussion

Careful modification of powder processing has enabled the manufacture of OTM materials coupled with more traditional ceramics that are of commercial interest. The sintering temperature and shrinkage of these advanced powders can be controlled to minimize residual stresses. FEM has accurately predicted the mechanical behavior of OTM tubes and suggested new strategies for increasing OTM robustness.

Thermodynamic modeling has suggested that under the majority of IGCC process conditions PSO1d is sufficiently stable. In addition, new compositions have been prepared that have improved stability in certain conditions that may arise in alternative IGCC process options.

C.2. Composite OTM Development Results and Discussion

High quality composite elements of PSO1d on PSO1d can be routinely prepared using a variety of processing methods. Composite elements are gas tight and have been tested for over 800 hours with no degradation in flux performance. Thin films of PSO1 have also been successfully deposited on substrates of other commercially viable materials. These samples have been tested for oxygen flux and have exhibited promising fluxes.

Methods for producing substrates with an improved porous structure have been developed. These substrates can be routinely prepared and thin films have been successfully deposited. These substrates have a permeation rate almost twice that of conventional substrates.

C.3. Manufacturing Development Results and Discussion

Initial fabrication of composite elements of PSO1d on PSO1d had a poor yield, with many samples exhibiting significant cracking. Modifications to the fabrication process were developed that has increased the yield of composite OTM elements, and the final strength of the sintered body. This process will be employed to fabricate elements up to three feet in length.

C.4. Process Development Results and Discussion

A composite tube comprising of PSO1 on a suitable substrate was tested in the high-pressure permeation tester at 900°C for over 300hrs. **The membrane was subjected to feed pressures in excess of 200psig and oxygen was produced at atmospheric pressure with a purity of greater than 95%.** Further experimentation on composite OTM elements with improved architecture are expected to yield higher flux performance. Post analysis of the composite OTM element has identified areas of improvement to meet commercial requirements.

C.5. O-1 Pilot Reactor Development Results and Discussion

An initial OTM element assembly design was selected to enable the construction of a multi-tube pilot reactor that will produce oxygen under process conditions simulating IGCC operation. The feed gas heating system was designed based on the results of process simulation models. The selected design is flexible and allows substantially different flow rates of the feed gas to be used during the experimental test program.

D. Conclusion

Good progress has been made in all tasks toward achieving the DOE-IGCC program objectives. In task 1, improved powder processing has enabled the production of elements that incorporate OTM films and substrates of commercial interest. In task 2, multiple fabrication processes have been developed that have been used to create stable OTM membranes and to deposit these on commercially relevant substrates. In task 3 modifications to the processing of OTM elements has led to an increase in the production yield of composite samples. In task 4**, oxygen has been produced at atmospheric pressure with a purity of greater than 95% in high pressure operation**. The work in task 5 has identified an OTM module design to meet the target oxygen production rate of the O1 pilot reactor.

E. References

1. Prasad, Ravi, "Ceramic Membrane Enabling Technology for Improved IGCC Efficiency" 1st Annual Technical Progress Report for US DOE Award No. DE-FC26-99FT40437, October 2000.