ENGINEERING DEVELOPMENT OF CERAMIC MEMBRANE REACTOR SYSTEMS FOR CONVERTING NATURAL GAS TO HYDROGEN AND SYNTHESIS GAS FOR LIQUID TRANSPORTAION FUELS

Monthly Report October 1999

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Contract Objectives

The objective of this contract is to research, develop and demonstrate a novel ceramic membrane reactor system for the low-cost conversion of natural gas to synthesis gas and hydrogen for liquid transportation fuels: the ITM Syngas process. Through an eight-year, three-phase program, the technology will be developed and scaled up to obtain the technical, engineering, operating and economic data necessary for the final step to full commercialization of the Gas-to-Liquids (GTL) conversion technology.

Summary of Activity

Task 1.1 Process Engineering and Economics

Task 1.1.1 Process Design and Engineering

Process development continued in evaluating potentially lower-cost process designs.

Task 1.1.2 Commercial-Scale Plant Economic Evaluation

The preliminary-level ITM Syngas process design and economics are being evaluated for two cases: an offshore GTL plant with 55 MMSCFD (60°F, dry basis) total associated gas feed and a land-based GTL plant with 500 MMSCFD (60°F, dry basis) total associated gas feed.

McDermott completed an interim report on the preliminary economic evaluation of offshore applications of the ITM Syngas process at two steam-to-carbon ratios. The results indicated that ITM technology has the potential to provide significant cost savings over currently available technology. Work was also initiated on an interim report to document the installed equipment costs for the 500 MMSCFD associated gas basis for Gulf of Mexico (GOM) and Alaskan North Slope (ANS) applications.

Task 1.1.3 PDU Systems Engineering

The design and engineering of the PDU was divided into several sections which will be individually quoted and engineered. The Design Hazards review documentation was begun, and the design of the PDU vessel was initiated by McDermott.

Task 1.2 Materials and Seals Development

Task 1.2.1 Materials Development

Subtask 1.2.1.1 Materials Characterization and Assessment

Material Characterization

Accelerated aging tests were begun at temperatures greater than the maximum process temperature and in various environments for a series of I4 compositions. These tests revealed that some of the I4 materials showed a slow decrease in oxygen flux and some decomposition in low P_{O2} atmospheres.

Mechanical Property Measurement

The mechanical properties of samples of various I4 compositions were measured in air at Penn State University.

Subtask 1.2.1.2 Powder Production and Test Sample Fabrication

Ceramatec fabricated various I4 samples for syngas testing at Air Products and Eltron Research, mechanical property measurement at Penn State University, and seal development at PNNL.

Subtask 1.2.1.3 Atmospheric Pressure Testing

The atmospheric pressure reactor at Air Products was used to test samples of additional modified I4 compositions.

At Eltron Research, atmospheric pressure testing of I4 membrane samples continued to evaluate the relative performance of reforming catalyst layers applied to the surface of the membrane. The conversion of the methane content of the feed stream to hydrogen and carbon monoxide was studied as a function of flow rate.

Subtask 1.2.1.4 Low DP (<300 psig) Testing

High-pressure reactors 1 and 2 at Eltron continued to be used to evaluate I4 membranes and seals supplied by Ceramatec. One test was run continuously at a pressure differential of 250 psia and 825°C under a reducing gas mixture for over 900 hours. At present, the oxygen flux based on the GC analysis of the product stream is higher than that measured by depletion of the air feed. A second test was run under similar conditions for >600 hours. The apparent oxygen flux based on the analysis of the syngas product increased with increasing air flow rate. A circumferential crack was observed in the tube at the end of the test.

Subtask 1.2.1.5 High DP (<500 psig) Testing

Pressure testing of the first Air Products 500 psig reactor was completed.

Task 1.2.2 Seals Development

Samples of ceramic-metal seals produced by Ceramatec and used in syngas tests at Eltron were analyzed at Air Products by SEM/EDS to determine any differences in microstructure that may be related to seal performance. Ceramic-ceramic seal development was also initiated at Ceramatec.

PNNL's modification of potential glass seal compositions resulted in increased thermal expansion coefficients, bringing several into good agreement with the target range. Their ability to bond ceramic and metal test samples was evaluated. A review of metallic braze product suppliers was carried out to identify a commercially available braze that could be used for a ceramic-metal joint.

Task 1.3 ITM Syngas Reactor Design and Fabrication

Task 1.3.1 ITM Syngas Reactor Design and Engineering

Subtask 1.3.1.1 Mechanical/Structural Design of Membranes and Seals

McDermott continued development of a planar membrane design and analysis of the thermal profile produced by the ITM Syngas reaction within a planar membrane configuration. Work was initiated on modeling the stress profiles in the ceramic-metal seal design fabricated by Ceramatec.

Subtask 1.3.1.2 Reaction Engineering and Kinetic Modeling

Chevron completed a summary of its reaction engineering and kinetics modeling work for tubular membrane configurations. A preliminary model for planar membrane configurations was proposed by Air Products and is being reviewed by Chevron and Norsk Hydro.

Subtask 1.3.1.3 Conceptual Reactor Vessel Engineering

The membrane and reactor vessel design and engineering of the shell and tube and planar reactor configurations were reviewed and evaluated at a meeting of Air Products, Ceramatec and McDermott. The planar reactor design was selected for further development. Alternative reactor vessel arrangements are being evaluated.

Task 1.3.2 ITM Syngas Membrane Fabrication

Subtask 1.3.2.1 Powder Production, Process Development and Scaleup

Ceramatec produced a total of 100 kg of I4 powder this month to investigate alternative I4 powder milling procedures as a prelude to scaling up to the production of larger batches of powder. Air Products investigated the correlation of the milling procedure with the purity of the calcined ITM Syngas powder.

Subtask 1.3.2.2 Membrane Fabrication Process Development

Ceramatec continued with the development of supported, thin-film membrane sample structures.