

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

Monthly Report November 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

Air Products, Chevron and Norsk Hydro initiated work on summary reports documenting the process design and engineering task.

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 began work on a report documenting both the offshore and land-based economic evaluations.

Task 1.1.3 PDU Systems Engineering

The design and engineering of the PDU continued. The initial DHR was prepared and will take place next month. A three-dimensional model of the initial reactor design is being fabricated in Plexiglas to assess issues such as syngas flow, membrane support, and assembly of the internals.

Task 1.2 Materials and Seals Development

Task 1.2.1 Materials Development

Subtask 1.2.1.1 Materials Characterization and Assessment

Material Characterization

Modified I4 material compositions demonstrated stable oxygen flux, and no decomposition in low P_{O2} atmospheres, in high-temperature accelerated aging tests.

Mechanical Property Measurement

The mechanical properties of samples of various I4 compositions continued to be measured at Penn State University. Modification of the material processing conditions resulted in a decrease in the observed non-linear load-time behavior.

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 second atmospheric pressure screening reactor at Air Products was completed. A slow decline in flux was observed in an atmospheric pressure test of an I4 membrane in an accelerated aging test carried out at temperatures higher than the maximum process temperature.

At Eltron Research, experiments were begun to study the performance of Ni catalysts supported on an I4 substrate. These data will be used to assess the effectiveness of a layer of reforming catalyst applied to the membrane surface.

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 of an I4 membrane continued for over 1450 hours at a pressure differential of 250 psia and 825°C under a reducing gas mixture. The oxygen flux based on the GC analysis of the product stream was higher than that measured by depletion of the air feed. An increase in the flow rate of air supplied at atmospheric pressure to the interior of the membrane increased the oxygen flux and altered the product composition. This was attributed to the air flow hydrodynamics. Another test of a tubular I4 membrane was begun at 900°C and 250 psia, and has run for 350 hours under process gas conditions. High methane conversions of >80% were observed.

Subtask 1.2.1.5 High DP (<500 psig) Testing

The first Air Products 500 psig reactor was operated at full process temperature and pressure, and the ORI (Operational Readiness Inspection) was completed.

Task 1.2.2 Seals Development

Air Products' SEM/EDS analysis of aged samples of ceramic-metal seals produced by Ceramatec showed inter-diffusion and the formation of oxides at the seal interface. At Eltron Research, ceramic-metal membrane seals fabricated by Ceramatec passed 1450 hours of testing at 825°C and 250 psig, and 350 hours of testing at 900°C and 250 psig.

Ceramatec continued the development of ceramic-ceramic seals and demonstrated a leak-tight I4 ceramic 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 completed an analysis of the air flow hydrodynamics in a planar membrane configuration. Detailed thermal and stress analyses of the gas flow arrangements are in progress for the planar membrane geometry, and the initial results showed reasonable temperature gradients.

Subtask 1.3.1.2 Reaction Engineering and Kinetic Modeling

Kinetic modeling work by Chevron and Air Products concentrated on the planar membrane reactor model.

Subtask 1.3.1.3 Conceptual Reactor Vessel Engineering

Initial planar membrane reactor vessel costs estimated by McDermott appear to be an improvement over those for the tubular reactor concept. The final costs will be incorporated into the economic evaluations.

Task 1.3.2 ITM Syngas Membrane Fabrication

Subtask 1.3.2.1 Powder Production, Process Development and Scaleup

Ceramatec continued the development of I4 powder production with an investigation of spray drying procedures. Approximately 10 kg of powder were sent to an external vendor for extrusion trials.

Subtask 1.3.2.2 Membrane Fabrication Process Development

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