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

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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 and Chevron considered additional process schemes for the ITM Syngas process in a GTL application with various reactor gas compositions.

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.

Phase 1 costs for the planar version of the ITM reactor vessel were incorporated by McDermott into the economic analysis.

Task 1.1.3 PDU Systems Engineering

The design and engineering of the PDU continued, with use of the Plexiglas model to assess the design and assembly of the reactor internals and the location and routing of instrumentation. McDermott continued the design and cost estimate of the reactor vessel.

Task 1.2 Materials and Seals Development

Task 1.2.1 Materials Development

Subtask 1.2.1.1 Materials Characterization and Assessment

Material Characterization

Non-stoichiometry measurements were made for selected I4 materials to provide data for modeling the oxygen flux in the ITM Syngas process.

Mechanical Property Measurement

The mechanical properties of samples of various I4 compositions continued to be measured 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

Atmospheric pressure testing of new I5 membrane compositions with improved high-temperature stability was continued at Air Products. The oxygen flux was confirmed to be stable over the test period at higher temperatures than the maximum process design temperature.

At Eltron Research, atmospheric pressure tests of tubular I4 membranes, and experiments to determine the performance of Ni catalysts supported on an I4 substrate, were continued.

Subtask 1.2.1.4 Low **DP** (<300 psig) Testing

The reactor used by Eltron Research for the 2400 hour test of the I4 membrane at 250 psia and 825° C, reported last month, was dismantled. Recalibration of the gas supply systems brought the oxygen flux into agreement within $\pm 25\%$, based on the GC analysis of the product stream and depletion of the air feed. Movement of the high-pressure reactors to new facilities created an opportunity to improve a number of support systems. These included changes to the gas delivery, product plumbing, analytical equipment, data acquisition and safety systems. The initial tests with each reactor will be conducted to confirm gas delivery and control systems. Reactors 3 and 4 are complete and under operation with I4 tubular membranes. Reactors 1 and 2 will be brought on-line when the mass flow controllers are recalibrated by the manufacturer.

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 shakedown of the system controls continued.

Task 1.2.2 Seals Development

Ceramatec prepared seal compositions and I4 ceramic test samples for further development of a ceramic-ceramic seal technology.

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 is constructing a finite element model of the commercial-size membrane. McDermott is also modeling alternative ceramic-metal seal concepts.

Subtask 1.3.1.2 Reaction Engineering and Kinetic Modeling

Kinetic modeling work by Chevron and Air Products continued for the planar membrane configuration. Thermodynamic data were assembled for all the species in the reaction system, including enthalpies of formation, free energies and heat capacities as a function of temperature.

Subtask 1.3.1.3 Conceptual Reactor Vessel Engineering

McDermott performed a global thermal analysis for the planar reactor configuration. The results were used as input to re-evaluate the thermal and structural analyses of the planar membrane design. Alternative vessel arrangement concepts were developed, and costs will be estimated.

Task 1.3.2 ITM Syngas Membrane Fabrication

Subtask 1.3.2.1 Powder Production, Process Development and Scaleup

An additional mill was installed at Ceramatec for processing I4 powders. Up to 60 kg per month of two different I4 compositions are planned to be produced for tubular membrane test samples and supported, thin-film membrane fabrication development.

Subtask 1.3.2.2 Membrane Fabrication Process Development

Additional samples of planar-supported, thin-film I4 membrane structures were fabricated by Ceramatec, and shown to be suitable for oxygen flux testing. Design of a preliminary membrane to be fabricated for the PDU was initiated.