Critical Component/Technology Gap in 21st Century Power Plant Gasification Based Poly-generation: Advanced Ceramic Membranes/Modules for Ultra Efficient H₂ Production/CO₂ Capture for Coal-Based Poly-generation Plants

DE-FE0031930

Carbon Management Project Review Meeting 8-16-2022

- Dr. Richard J. Ciora, Jr., Media and Process Technology Inc., Pittsburgh, PA
- Professor Theo Tsotsis, University of Southern California, Los Angeles, CA



MPT TECHNOLOGY BACKGROUND

Project Overview

<u>Program:</u> DOE/FE "Critical Components for the 21st Century Power Plant" <u>Funding:</u> Project Budget/Cost Share: \$2.38M (DOE: \$1.91M; Cost Share: \$0.47M) <u>Overall Project Performance Dates:</u> October 1, 2020 - September 30, 2023 (36 months) <u>Project Participants:</u>

- Media and Process Technology Inc...Membrane manufacturer/supplier and technology developer (POC: Dr. Richard Ciora)
- University of Southern California....Membrane and system modeling (POC: Professor Theo Tsotsis)

Overall Project Objectives:

Perform R&D to enable emerging inorganic membrane technology in Poly-generation based precombustion CO_2 capture.

- (i) Develop & fabricate a full ceramic multiple tube bundle w/ permeate purge capability.
- (ii) Fabricate multiple bundle housing/module as a pre-commercial scale unit.



MPT TECHNOLOGY BACKGROUND

Poly-generation: Inorganic Membrane Technology Role





Project Objectives and Technical Program Summary

<u>Primary Objective</u>

Develop a permeate sweep/purge capable full ceramic multiple tube membrane bundle and multi-bundle module.

- ✓ This capability represents a Critical Technology Gap for advanced inorganic membranes in pre-combustion CO₂ capture.
- \checkmark This capability is required to achieve the >30% COE cost savings target over baseline.
- ✓ Target operating conditions of 200 to >350°C and at up to 800 psig.

Technical Program Summary

- > Develop and fabricate a permeate purgeable multiple tube bundle
- > Design and fabricate a multiple bundle housing with appropriate seals
- Conduct a range of challenge tests to demonstrate bundle/housing stability
- Conduct long term performance stability testing at the target operating conditions
- Develop CFD model to predict membrane performance and inform module configuration
- Update the DSMP TEA for Poly-generation



M&P TECHNOLOGY BACKGROUND





Permeate Purgeable; Multiple Bundle Housing and Seals





Project Structure...Team Member Roles and Tasks





Concepts and Milestones: BP1

Task 2. Dual End Bundle DevelopmentMilestone d:Dense tube bundles (9/21).Milestone e:Porous tube bundles (3/22).





Project Activities

- ► 1.5", 2" and 3" diameter ✓
- T/C stability to 450° C \checkmark
- Constrained/mechanically loaded
 T/C stability (in housing) ✓
- 4" Dual End Bundle

Task 3. Housing and Seal DevelopmentMilestone f:Single bundle housing (6/21).Milestone g:Multiple bundle housing. (3/22)



Task 4. Mathematical Model DevelopmentMilestone h:Single bundle housing.Milestone i:Multiple bundle housing.





Project Activities

- Radial and face seal designs
- Multiple bundles in series \checkmark
- Permeate sweep capable ✓
- Continue long term stability testing.

Project Activities

- ► Feed flow distribution, multiple tube ✓
- Incorporate permeation/separation
- Multiple bundle in series/parallel
- Module layout optimization.



Concepts and Milestones: BP2

Task 5. Dual End Pd-alloy and/or CMS Membrane BundlesMilestone j: Minimum target is 4x bundles with gasseparation and permeate purge capability.



Project Activities

- Fabricate single tube Pd-alloy and CMS membranes.
- Fabricate multiple dual end membrane bundles
- Conduct mixed gas performance testing

Task 6. Evaluate the operating envelope of a two bundle housing. <u>Milestone k:</u> Complete shakedown and permeance testing in permeate sweep gas mode of two bundle module.



Project Activities

Evaluation of two Pd-alloy and/or CMS membrane bundles in mixed gas testing with permeate sweep. Task 7. Test Prototype Full Scale MultipleBundle Housing.Milestone I:Complete multiple bundle test.



Project Activities

- Develop and test the membrane module with parallel and series configured bundle array.
- Complete testing in the base modular unit (target is six bundle array)

Task 8. Update the TEA.Milestone m:Update the TEA with newmodule design and process configuration.

Project Activities

 Update Polygeneration PFD and TEA with purge capable bundles



Success Criteria. BP1

Success Criteria	Description	Status
#1	Successful completion of Tasks 1 through Subtask 4.2	All tasks complete up to subtask 4.2 CFD model sensitivity/optimization.
#2	Successful fabrication of three to four multiple tube full ceramic bundles in the dual end configuration at a minimum diameter of at least 1.5" and (stretch) target diameter of 4" at a length between 20(18)" to 38".	<i>Success.</i> 43 dense and porous tube bundles successfully fabricated at 1.5", 2", and 3" diameter at 18 to 38" length.
#3	Bundles are qualified by meeting a target leak rate of <1 GPU of nitrogen and minimum compression test up to 400psig at 250°C.	<i>Success.</i> Multiple bundles demonstrated to be leak free under a wide range of thermal and pressure cycle challenge testing. No bundle failure in over 1,000 cycles.
#4	Successful fabrication of the first operational housing for single membrane bundles in the proposed configuration with negligible leaking (i.e., N_2 permeance < 1 to 5 GPU) or physical failure during the test at temperatures up to 450°C and pressures up to 800psig.	<i>Success.</i> Multiple bundles in housings with demonstrated leak rates <<1 GPU under testing up to these operating conditions.
#5	CFD modeling of the dual ended bundles in U-bend connection (centerline permeate takeoff) is completed and recommendations for feed flow distributors (baffles, etc.) are incorporated.	<i>Success.</i> CFD model complete with multiple tube bundle with side entry and permeate sweep. Sensitivity/optimization analysis underway for feed flow distributors (baffles).



BP1 Success Criteria #2

Task 2. Fabricate Permeate Purgeable Bundles

1.5": 18 x 5.7mm Dense/Porous Tube w/ Centerline Permeate Takeoff

"First-of-its-kind"

2": 12 x 8.6mm Tube Dense/Porous Tube w/Centerline Permeate Takeoff

3": 4x 8.6mm Dense Tube w/Centerline Permeate Takeoff

3": 1x 5.7mm Porous Tube w/Centerline Permeate Takeoff (36-tube; 0.6m² surface area at 38")

Progress Summary

- 1. <u>1.5" to 3" bundles</u>: Routine.
- 2. <u>Centerline Permeate</u> <u>Takeoff:</u> Routine.
- 3. <u>Porous Tube Bundles:</u> Successful, routine.
- 4. <u>4" bundles</u>: Should be straightforward; solved the 3" bundle fabrication problem.

Bundle Challenge Testing

	Thermal Cy	cling, "Unlo	aded"			Thermal Cycling, in Housing		Housing	Pressure Cycling, in Housing		
Membrane ID	250°C	400°C	450°C	Membrane ID	Housing/Seal	250°C	400°C	450°C	250°C	400°C	450°C
PG1.5-PTA-1	13	(101) 160	25	PG1.5-PTA-1	Single Face, Metal	28	27	24		6	6
PG1.5-PT-2	0	(114) 173	0	PG1.5-PT-2							
PG1.5-PT-3	0	(91) 150	23	PG1.5-PT-3							
PG1.5-DT-2	8	63	2	PG1.5-DT-2							
PG1.5-DT-3	8	62	23	PG1.5-DT-3							
PG1.5-DT-6	0	0	34	PG1.5-DT-6							
PG1.5-DT-7	0	0	0	PG1.5-DT-7	Radial, Graphite	30	37	24			
PG1.5-DT-8	1	6	0	PG1.5-DT-8	Radial, Graphite	10	0	9	8	6	
PG1.5-DT-9	0	7	0	PG1.5-DT-9	Dual Face, Metal	10	3	7		3	
(PG1.5-DT-9)	0	0	0	(PG1.5-DT-9)	Single Face, Oring	10			18		
PG1.5-DT-10	0	4	0	PG1.5-DT-10	Dual Face, Metal	9	3	7		3	
PG1.5-DT-11	0	13	12	PG1.5-DT-11							
PG1.5-DT-12	0	12	24	PG1.5-DT-12	Dual Face, Oring				67		
PG1.5-DT-14	0	0	0	PG1.5-DT-14	Dual Face, Oring				39		
PG1.5-DT-15	0	0	0	PG1.5-DT-15	Dual Face, Oring				20		
PG1.5-DT-16	0	0	0	PG1.5-DT-16	Dual Face, Oring				75		
PG1.5-DT-6	0	34	16	PG1.5-DT-6							
PG1.5-DT-13	0	0	0	PG1.5-DT-13							
PG1.75-DT-1	0	27	0	PG1.75-DT-1	Radial, Graphite		8		9	(27) 62	
PG1.75-DT-2	0	24	0	PG1.75-DT-2							
PG1.75-DT-3	0	10	0	PG1.75-DT-3	Dual Face, Oring				6		
PG1.75-DT-4	0	8	0	PG1.75-DT-4	Dual Face, Oring				6		
PG1.75-DT-7	0	0	0	PG1.75-DT-7	Dual Face, Oring				29		
PG1.75-DT-8	0	0	0	PG1.75-DT-8	Dual Face, Oring				3		
PG1.75-DT-9	0	0	0	PG1.75-DT-9	Dual Face, Oring				26		
PG1.75-DT-10	0	0	0	PG1.75-DT-10							
PG1.75-DT-11	0	0	0	PG1.75-DT-11							
PG1.75-DT-12	0	0	0	PG1.75-DT-12	Dual Face, Oring				(0) 53		
PG1.75-DT-13	0	0	0	PG1.75-DT-13	Dual Face, Oring				(0) 53		
PG1.75-DT-14	5	(3) 6	0	PG1.75-DT-14							
PG1.75-PT-2	5	(3) 6	0	PG1.75-PT-2							
PG2-PT-1	0	0	33	PG2-PT-1	Radial, Graphite		9	24		4	
PG <mark>2-PT-</mark> 2	0	6	8	PG <mark>2-PT-</mark> 2							
PG <mark>2-PT</mark> -3	0	0	8	PG <mark>2-PT-</mark> 3	Radial, Graphite			22		15	
PG2-PT-4	0	0	8	PG <mark>2-PT-</mark> 4							
PG <mark>2</mark> -DT-1	0	4	22	PG2-DT-1							
PG <mark>2</mark> -DT-2	0	28	0	PG2-DT-2							
PG <mark>2</mark> -DT-7	0	0	0	PG2-DT-7	Radial, Graphite	21	12			17	
PG3-DT-1	12	8	0	PG3-DT-1							
PG3-DT-2	4	58	0	PG3-DT-2							
PG3-PT-1	0	(64) 125	0	PG3-PT-1							
PG3.5-DT-5	10	58	0	PG3.5-DT-5							
PG3.5-DT-6	2	0	0	PG3.5-DT-6							
Totals	68	1,052	238		Totals	118	99	117	412	116	6

BP1 Success Criteria #3

Key PGxx-yy-ID# xx: Bundle Diameter yy: DT = Dense Tube PT = Porous Tube

Results (7-2022)

Thermal Challenge Testing:

- 1. 43 Successful Dual End Bundles
- 2. >1,300 T/C unloaded
- 3. >300 T/C loaded housing
- 4. >500 T/C loaded, pressurized

Focus During BP2

Long term mechanical stability. Pressure loaded housing + seal. Larger Bundles. 3" Porous Tube Larger Bundles. 4" Porous Tube

Technical Approach: BP1 *Task 6.* Evaluate operating envelope of two bundle housing. Dual End Full Bundle Housing (Single Bundle and Dual Bundle) in Face Seal Configuration Bundles Stationary High Load

Temperature Sealing Fixture

2" Dual End Bundle Radial Seal Housing



1.5" Dual End Bundle Radial Seal Housing

Face Seal





Spring

<u>3" Dual End Bundle</u> Radial/Face Seal Housing Testing Beginning 6/2022

Face Seal



Task 2. Performance Test Permeate Purgeable Bundles

High Pressure Performance Testing. Radial Seal Modules





BP1 Success Criteria #4



Task 2. Performance Test Permeate Purgeable Bundles

Two-in-Series Dual End Bundle Face Seal Housing

High Pressure Performance Testing





BP1 Success Criteria #4





Task 3. Purge Capable Module and Seals Design and Fabrication

BP1 Success Criteria #4

Summary of Dual End Bundle Challenge Testing

Leak Rate Targets: Project < 1 GPU; MPT < 0.35 GPU

Housing	Bundle ID [-]	Packing [-]	Time On-line [days]	Temperature Max [°C]	Pressure Max [psig]	Leak Rate [GPU]
1.5"	DT-12, -16	Face; O-ring	5	250	400	0.017
1.5"	DT-14, -16	Face; O-ring	19	250	800	0.009
1.5"	DT-14, -15	Face; O-ring	101	250	800	0
1.5"	DT-8	Radial; Graphite	6	400	800	0.24
2" Nominal	DT-3, -4	Face; O-ring	8	250	400	0.009
2" Nominal	DT-12, -13	Face; O-ring	53	250	400	< 0.01
2" Nominal	DT-1	Radial; Graphite	28	350	800	0.0165
	DT-1	Radial; Graphite	4	400	400	0.73
2" Nominal	DT-2 (single end)	Face; Metal	54	400	800	0.011
2" True	DT-7	Radial; Graphite	4	250	400	0
	DT-7	Radial; Graphite	5	400	550	0.085
2" True	Dense Blank Puck	Radial; Graphite	6	400	400	0
3"	Dense Blank Puck	Radial; Graphite	8	400	400	0



Success Criteria #5

Task 4. Conduct Mathematical and CFD Modeling

COMSOL Model Predictions

Concentration Profiles and Performance Predictions in Side Entry Housing





A. Side Entry Module Predictions						
Tube #	Tube Flow	He Purity				
[-]	[x10 ⁻⁷ kg/s]	[%]				
1	9.26	97.03				
2	9.31	97.06				
3	9.07	96.91				
4	8.94	96.84				
5	8.67	96.66				

B. Side Entry vs. Parallel Flow Predictions					
Model	Permeate	Permeate			
Configuration	He Recovery	He Purity			
	[%]	[%]			
Parallel Flow	76.1	96.9			
Side Entry	75.9	96.87			



Task 5. CMS and Pd (alloy) Tube and Bundle Fabrication

Pd "Low" Quality Tubes From Stock Inventory: 31

Tube ID	Temp	H ₂	H_2/N_2
[-]	[°C]	[GPU]	[-]
21	350	1,216	1,171
24	350	748	1,466
25	350	768	468
37	350	1,032	694
40	350	1,127	1,396
74	350	1,053	1,499
76	350	1,046	953
77	350	975	1,171
79	0	0	0
81	350	1,126	1,438
82	350	761	582
88	350	1,243	2,581
4	350	1,053	318
30	350	802	737
44	350	1,358	589
83	350	1,309	797
86	350	1,153	602
23	350 1,029		309
29	350 1,026		1,209
22	350	1,095	533
31	350	1,255	1,116
	Average	1,008	935

Pd "High" Quality Tubes BP2 Production Inventory: 42

Tube ID	Temp	H ₂	H_2/N_2
[-]	[°C]	[GPU]	[-]
1	350	3,940	6,034
2	350	4,007	37,336
3	350	4,758	9,653
4	350	4,536	40,794
5	350	3,505	29,886
6	350	3,520	31,881
7	350	4,599	180,337
8	350	4,368	110,211
9	350	3,376	35,889
10	350	3,702	15,011
11	350	3,513	29,914
12	350	3,257	8,905
13	350	3,257	106,926
14	350	3,727	335,602
15	350	3,551	47,749
16	350	3,621	177,892
26	350	3,059	18,277
27	350	4,988	198,003
	Average	3,849	78,906

CMS Dual End Tubes BP2 Production Inventory: 32

Tube ID	Temp	He	H_2/N_2	
[-]	[°C]	[GPU]	[-]	
DB04	250	363	231	
DB05	250	292	120	
DB06	250	389	360	
DB09	250	244	112	
DB11	250	243	324	
DB13	250	223	204	
DB15	250	327	246	
DB16	250	358	278	
DB17	250	433	200	
DB18	250	238	723	
DB19	250	421	169	
DB20	250	239	593	
DB22	250	316	209	
DB29	250	431	405	
DB25	250	697	269	
DB26	250	446	358	
DB31	250	278	557	
DB32	250	401	706	
DB27	250	382	346	
	Average	384	349	



Task 5. CMS and Pd (alloy) Tube and Bundle Fabrication

Pd Bundle Testing Bundle ID: PdB6T.01 6-Tube; 18" overall length; (3 full length parts cut in half)

Tube ID	Temp	H ₂	N ₂	H_2/N_2	Comment
[-]	[°C]	[GPU]	[GPU]	[-]	[-]
-24	350	748	0.51	1,466	
-40	350	1,127	0.81	1,396	
-74	350	1,053	0.70	1,499	
Bundle Pd	6 T. 01				
Day 1	RT	N/A	0.314		
Day 1	350	813.00	3.734		
Day 1	350	567	4.61	123	
Day 2	RT	N/A	0.49		Repack
Day 2	250	N/A	0.49		
Day 2	350	893	0.61	1473	
Day 5	450	327	0.77	425	
Day 5	350	983	0.91	1082	Air purge.
Day 6	350	726	1.02	712	
Day 8	350	1,056	0.83	1265	Power Outage.
Day 12	350	1,214	1.57	773	Air purge.
Day 13	350	N/A	1.68	N/A	
Day 20	350	1,366	2.08	657	Power Outage.
Day 28	350	N/A	1.09	N/A	Goto Mixed Gas





Run

ID

[#]

23

31

39

43

45

112

114

116

Bundle Testing. Modified feed inlet.

Single Tube Testing

Part

ID

[#]

31A

31A

31A

31A

31A

PdB6T-01

PdB6T-01

PdB6T-01

Bundle Testing

Feed

Rate

[cc/min]

352

347

573

643

492

4,560

4.611

4.501

Feed

Pressure

[psig]

43.5

41.0

37.5

35.5

42.0

46.0

47.0

47.8

Technical Approach: BP2

Task 5. CMS and Pd (alloy) Tube and Bui A. Single Tube: Excellent

Actual Permeate

Rate

[cc/min]

111

139

144

223

206

664

664

655

Predicted

Permeate

Rate/

[cc/min]

117

137

148

232

202

937

966

1.098

Pd Membrane Performance Mixed Gas Testing Single Tube and 6-tube Bundle with Permeate Sweep; Temperature: 350°C; Feed: 50/50 H₂/CO₂; Sweep Gas: N₂

Sweep

Rate

[cc/min]

0

70

0

170

170

0

0

158

A. Single Tube: Excellent agreement between mixed gas performance and "ideal" simulation based upon pure component permeances.

B. Permeate Sweep: Substantially improves membrane productivity.

C. 6-tube Bundle: Poor agreement between mixed gas performance and "ideal" simulation based upon pure component permeances. See COMSOL simulation in follow-up slide.







Task 4. Conduct Mathematical and CFD Modeling



Task 8. PFD and TEA Update





Summary. BP1

Summary of Project Status and Successes

- 1. <u>Dual End Bundle Fabrication</u>. Successfully fabricated 43 dual end tube side (permeate) purgeable dense and porous tube bundles at 1.5", 2", and 3" diameters ("first-of-its-kind").
- 2. <u>Bundle Challenge Testing (free expansion)</u>. Demonstrated bundles mechanically stable in thermal cycle challenge testing to 450°C in unloaded free-expansion mode.
- 3. <u>Bundle Challenge Testing (mechanically loaded in housing)</u>. Demonstrated bundles mechanically stable in thermal cycle challenge testing to 450°C in various housing styles (radial and face seal).
- 4. <u>Bundle Leak Performance Testing (mechanically loaded in housing at high pressure).</u> Demonstrated bundle seal leak rates well below the 1 GPU target in radial and face seal style housings at temperatures to 400°C.
- 5. <u>Multiple Bundle Housing with Permeate Purge Capability.</u> Demonstrated a two bundle housing with permeate purge capability ("first-of-its-kind").
- 6. <u>Developed a COMSOL based CFD Model for Membrane Performance Prediction.</u> Successfully predicted the mixed gas performance of an 18-tube membrane bundle. Includes side entry/exit of feed/reject and permeate side sweep.



Summary. BP2

Next Step

- 1. <u>Pd and CMS Tube fabrication underway.</u> High performance H_2 -selective membranes. ~150 of each required.
- 2. <u>First dual end bundles fabricated from gas separation tubes.</u> First set of 3- and 6-tube dual end bundle completed in Pd and CMS.
 - a. Project focus: 19-tube bundles in multiple bundle housing.
 - b. Stretch Target: Prototype 3" and 4" bundle scaleup with long-term challenge testing.
- 3. <u>Bundle Challenge Testing (mechanically loaded in housing; high pressure and temperature).</u> Mechanical and performance stability with dual end Pd and CMS bundles underway.
- 4. <u>Multiple Bundle Housing with Permeate Purge Capability.</u> Project target: Demonstrate the base 6-bundle housing with 2-series/3-parallel layout.
- 5. <u>COMSOL CFD Model for Membrane Performance Prediction</u>. Optimization of the bundle layout and housing design.

Questions?