Roll-to-Roll Manufacturing of Solid Oxide Fuel Cells

(TCF-20-20119)

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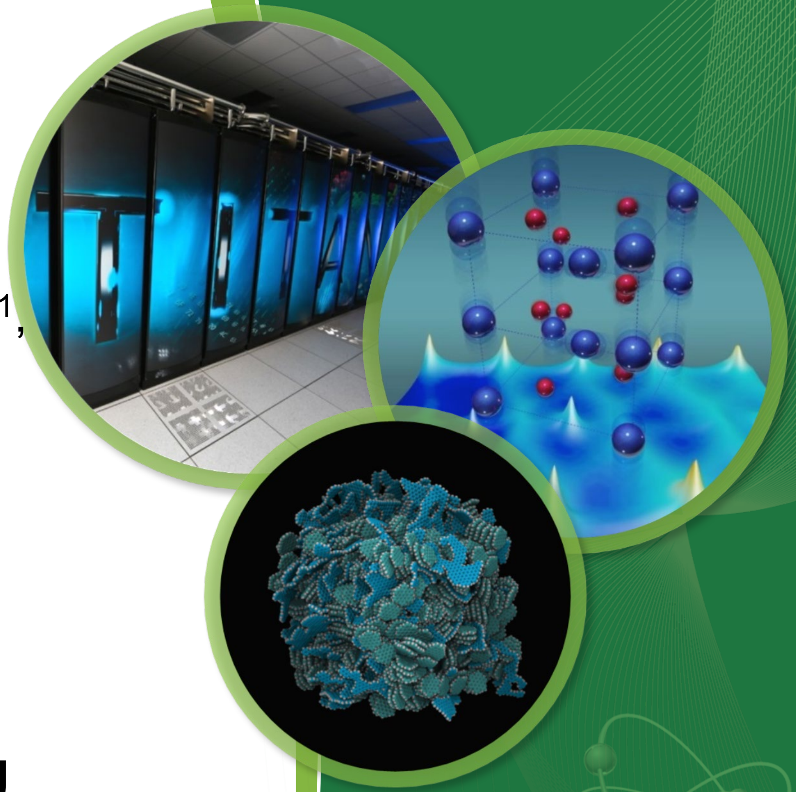


2024 FECM/NETL Spring R&D Project Review Meeting

Reversible Solid Oxide Fuel Cells

Thursday April 25, 2024

12:15 p.m. - 12:45 p.m.

Overview

Timeline and Budget

- Project Start Date: February 17th, 2022
- Project End Date: February 16th, 2025
- Total Project Budget: $ 2,400,000
  - Total DOE Share: $ 1,200,000
  - Total Cost Share: $ 1,200,000
  - Total DOE Funds Spent*: $ 236,473
  - Total Cost Share Funds Spent*: $ 37,346

* As of 03/01/2024

Barriers & Targets

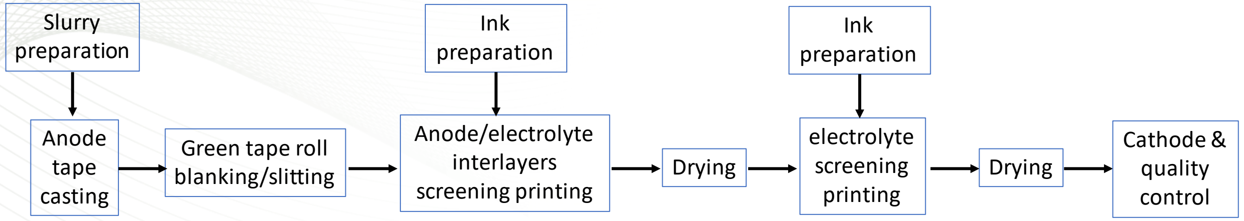
- Optimize the lamination process via calendering.
- Quantity scale-up of the lamination process to demonstrate > 10 ft of EEA.
- Further increase EEA throughput via slot-die coating and demonstrate > 5 m/min coating of ASL (10X increase vs tape casting).
- Minimize the anode supported layer (ASL) thickness and reduce material cost by >10%.

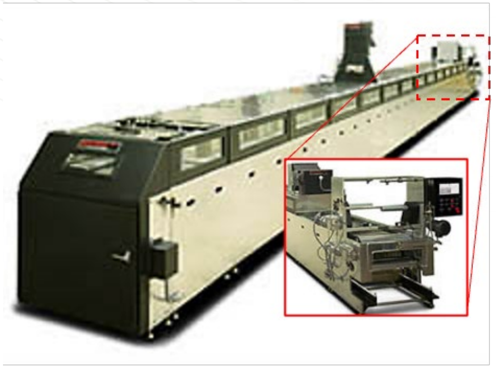
Partners

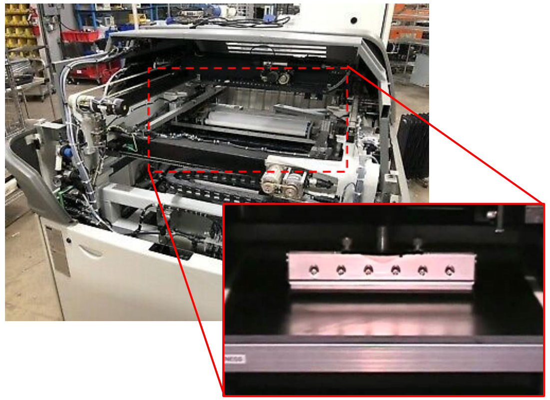
- ORNL (PI)
- Redox Power Systems

SOFC/SOEC manufacturing involves many steps and the manufacturing cost highly depends on the scale

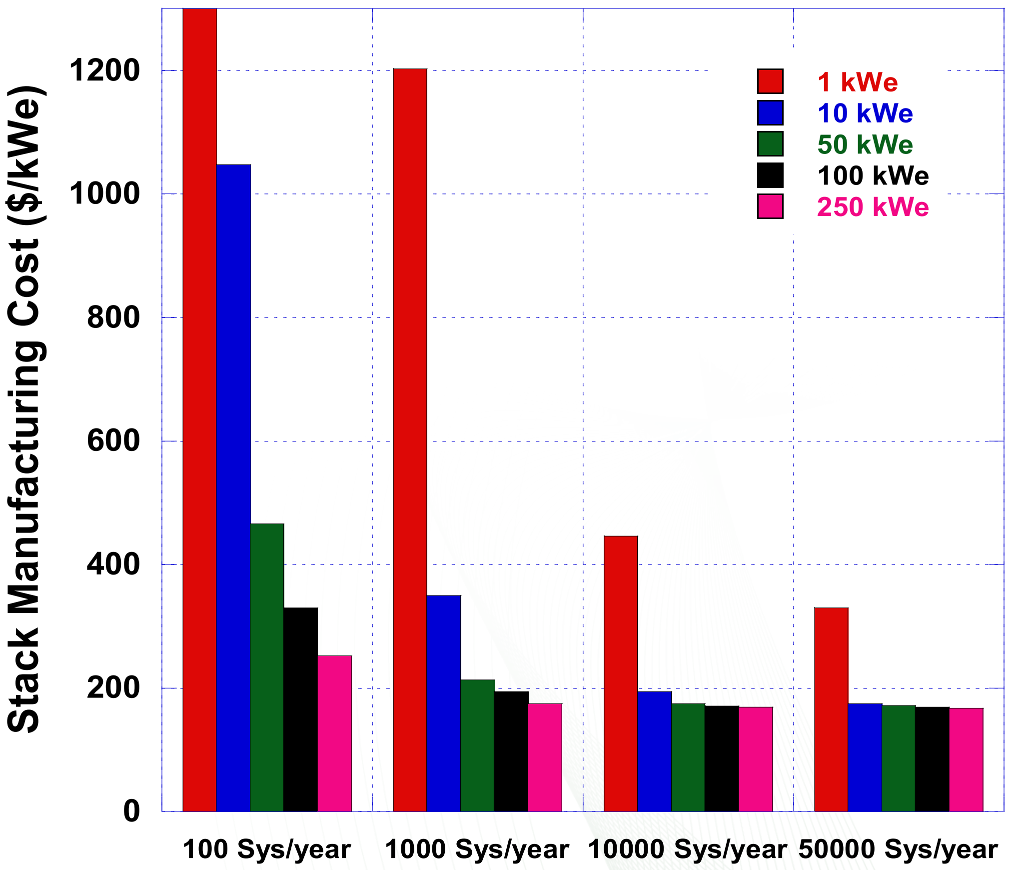
An example of an EEA manufacturing process







Stack manufacturing cost variation with system size.



Electrode and electrolyte were coated via tape cast and screening printing in sheet-to-sheet manner

- Low manufacturing volume
- High manufacturing cost
- Conventional method throughput is typically 5-8 min/laminate

Manufacturing cost can be significantly reduced by increasing manufacturing volume and system size

R. Scataglini, et. al., "A total cost of ownership model for solid oxide fuel cells in combined heat and power and power-only applications", Lawrence Berkeley National Laboratory, December 2015

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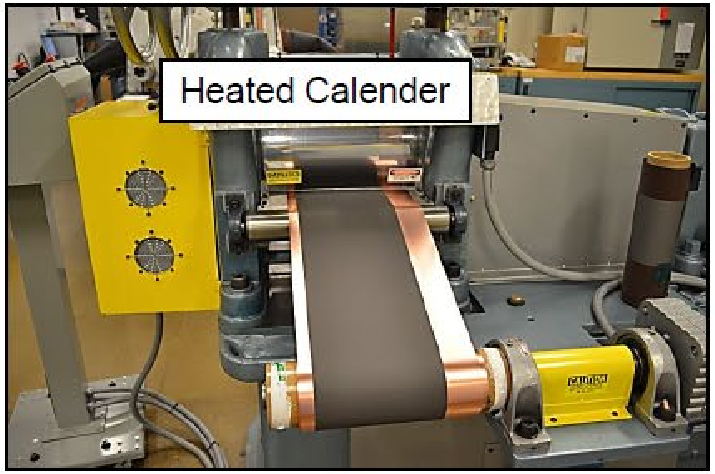
Roll-to-roll manufacturing capabilities at BMF

Battery electrode fabrication



- Planetary mixer
- ½ gallon

- Single and double sided
- Continuous and intermittent coating
- Capable of simultaneous multi-layer coating
- Up to 15 ft/min

- 9 drying zone including 2 IR zones

- 80,000 lb separating force
- Temperature up to 250^{o}C
- Up to 15 m/min

Roll-to-roll manufacturing capabilities at BMF

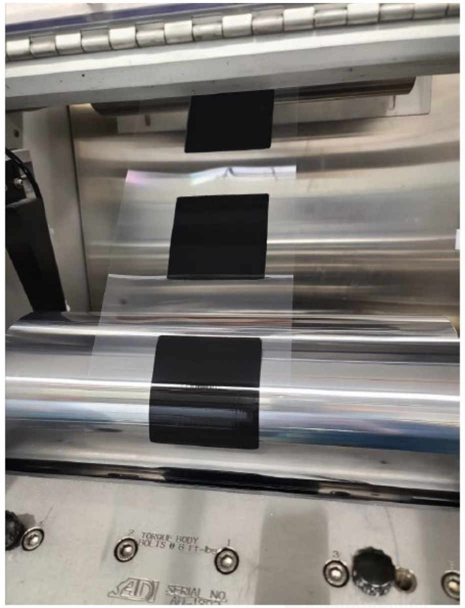
Thin film mixer

Bench top slot-die coater

Pilot scale slot-die coater

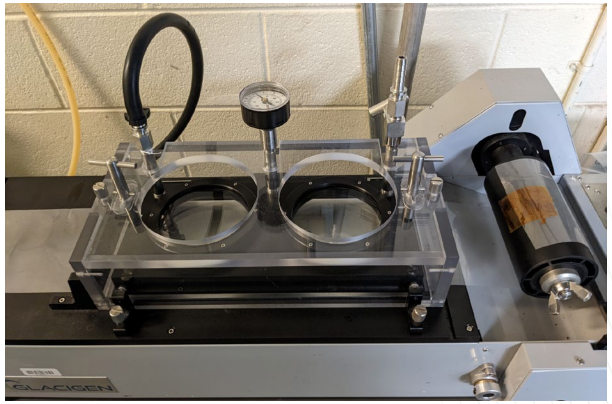
Electron beam curing

Patch coating

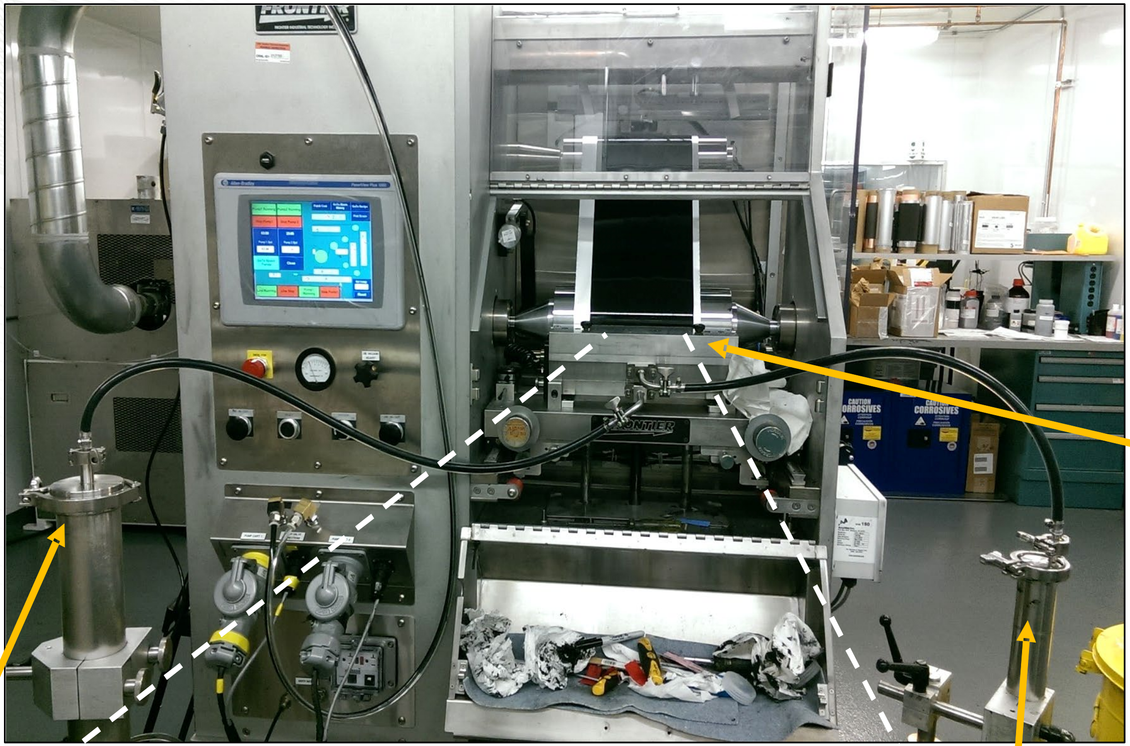


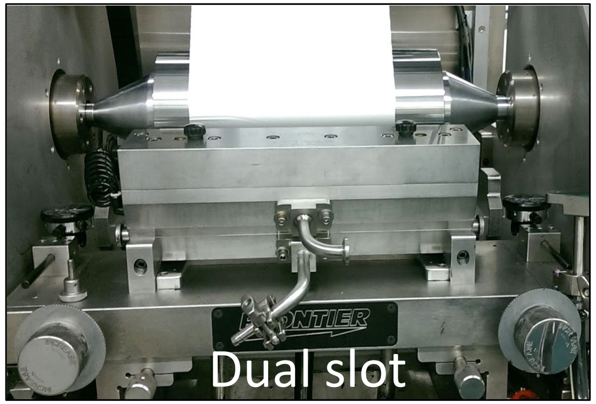
Heated calender

Two roll-to-roll freeze tape casters



Pilot scale slot-die coater at ORNL

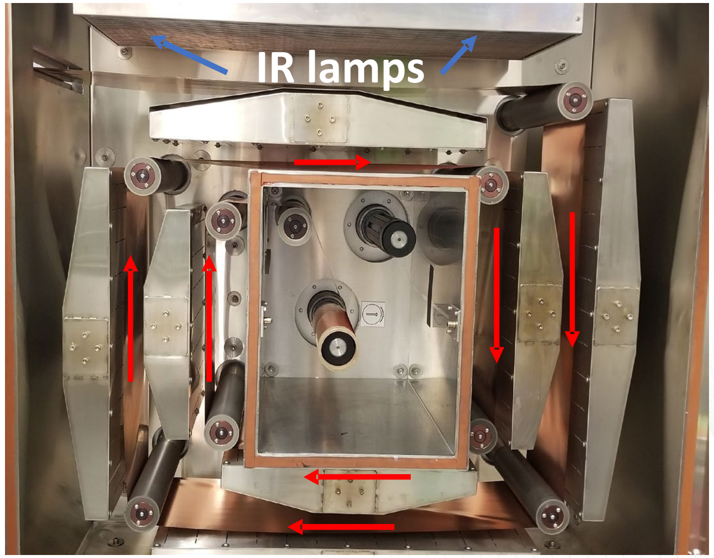




Slot Die

Slurry for Bottom Coating

Slurry for Top Coating



- Slot-die coating is a high precision and high throughput method to produce coating
- Dual slot-die can coat two layers simultaneously
- 9 drying zones provide flexibility to optimize drying protocols

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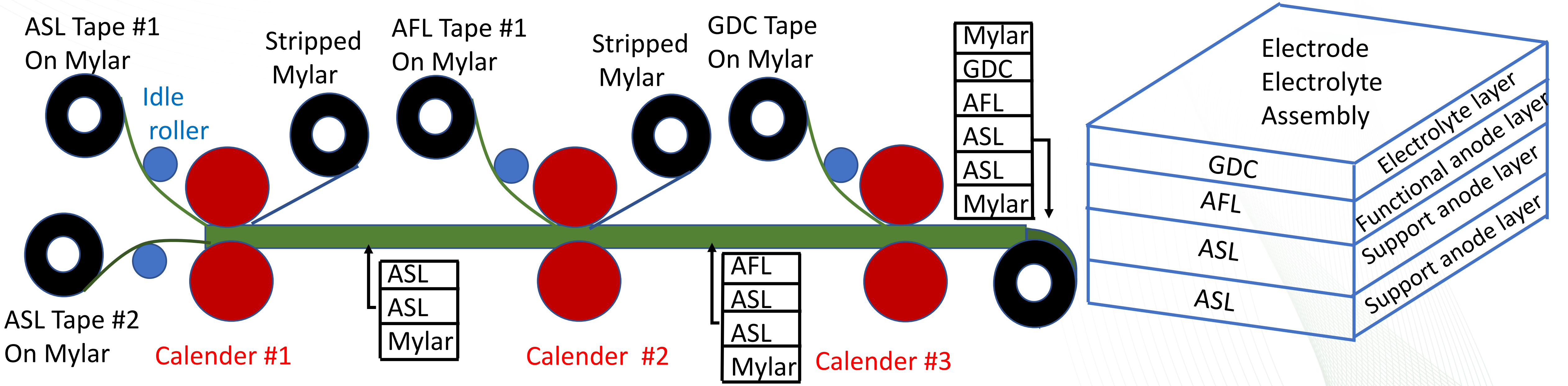
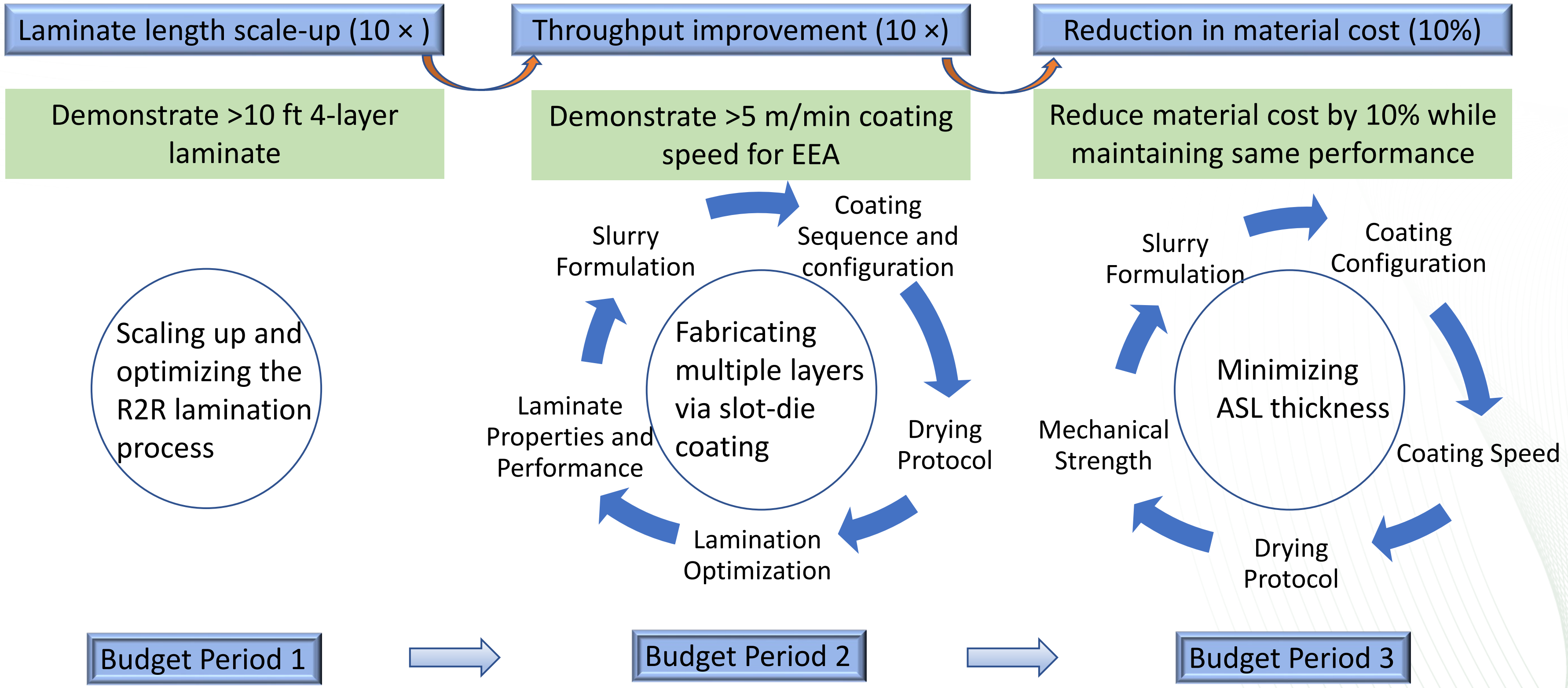
Roll-to-roll lamination process can produce electrode electrolyte assembly with high throughput

Goal: To further develop a high-volume electrode electrolyte assembly (EEA) production capability to significantly increase throughput of SOFC manufacture and reduce the manufacture cost by 30%.



Deliverables:

- **Budget Period 1:** Optimization of the lamination conditions and demonstration of >10 ft 4-layer EEA; sintering the EEA, characterizing the EEA morphology and properties, and testing the cell performance at ORNL and Redox.
- **Budget Period 2:** Fabricating multiple layers via slot-die coating, demonstration of >5 m/min in coating ASL, laminating them into EEA to reduce interfacial resistance, testing the cell performance and characterizing the laminate properties at ORNL & Redox.
- **Budget Period 3:** Further reducing SOFC cost by minimizing ASL thickness and enabling >10% material cost reduction



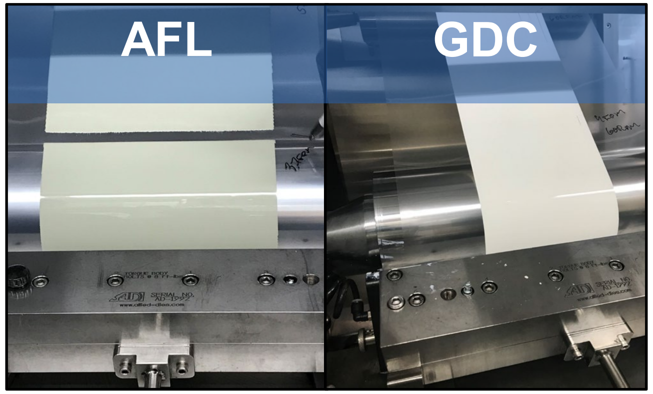
Project Milestones

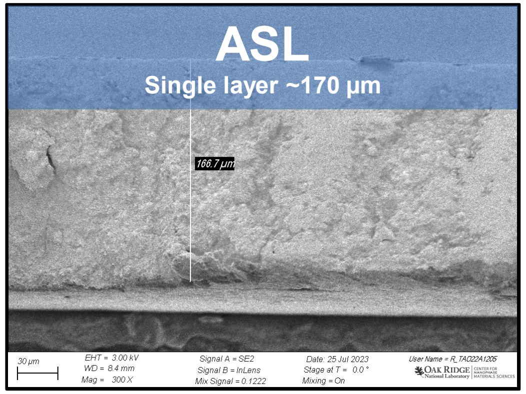
Phase	Timeline (Month)	Description (ORNL/Redox)
Phase 1 Optimization of the lamination conditions and demonstration of >10 ft 4-layer EEA; sintering the EEA, characterizing the EEA morphology and properties, and testing the cell performance at ORNL and Redox.	Task 1.1 (1) ✓ Task 1.2 (2-10) ✓ Task 1.3 (6-12) ✓	1.1 - Fabricate the ASL, AFL and GDC tapes via tape casting (Redox). 1.2 - Optimize the lamination conditions to produce >10 ft 4-layer EEA (ORNL). 1.3 - Sinter and characterize the EEA and test cell performance (ORNL & Redox).
Phase 2 Fabricating multiple layers via slot-die coating, demonstration of >5 m/min in coating ASL, laminating them into EEA to reduce interfacial resistance, testing the cell performance and characterizing the laminate properties at ORNL & Redox.	Task 2.1 (13-19) ✓ Task 2.2 (15-22) ✓ Task 2.3 (20-24)	2.1 - Fabricate individual layers and EEA via slot-die coating (> 5 m/min) (ORNL). 2.2 - Laminate the layers into EEA (ORNL). 2.3 - Sinter and characterize the EEA and test cell performance (ORNL).
Phase 3 Further reducing SOFC cost by minimizing ASL thickness and enabling >10% material cost reduction and further increase coating speed.	Task 3.1 (13-19) ✓ Task 3.2 (15-22) Task 3.3 (20-24)	3.1 - Coat ASL at various thickness (ORNL). 3.2 - Sinter ASL, characterize morphology & mechanical strength (ORNL & Redox). 3.3 - Produce EEA with thin ASL and test cell performance (ORNL & Redox).
All Budget Periods	Milestone 4 (1-36)	4 - Deployment, dissemination (ORNL & Redox).

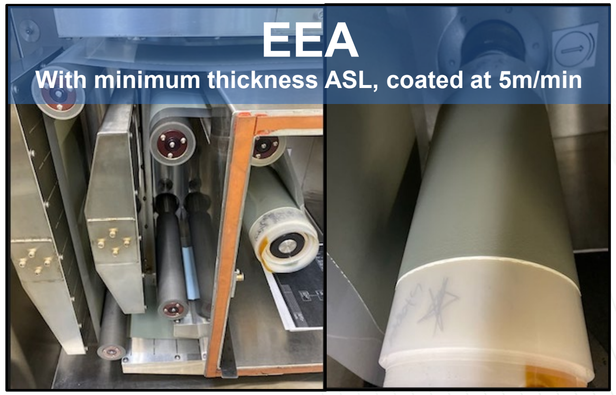
Previous Accomplishments:

(Project started in May 2022)

- Summary of the accomplishments prior to FY24:
  - Slurry formulation with rheology study to evaluate slurry processing conditions and improved coating behaviors and dry coating quality.
  - Identified coating and lamination conditions of the AFL, ASL and GDC layers.
  - Coated EEA of ASL/AFL/GDC at >5m/min using slot die coating.
  - Optimized ASL thickness
  - Evaluation of ASL thickness on processing, quality and mechanical behaviors in fabricated the EEA.







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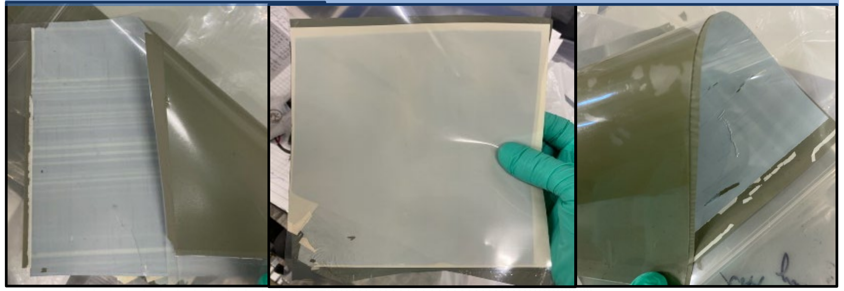
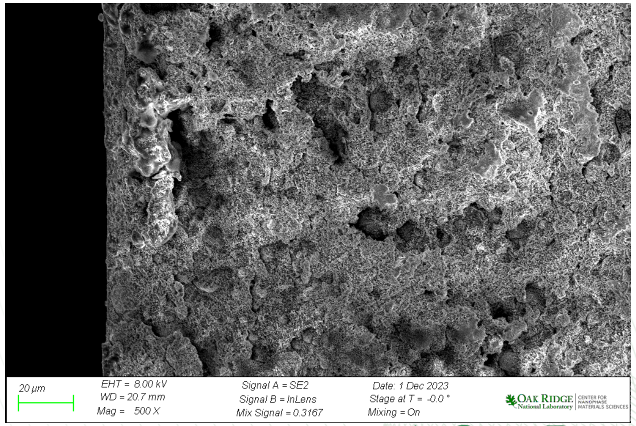
FY24 Accomplishment

- Fabricated EEA from coated layers and established lamination window for processing EEA with excellent interlayer contact using the 4-High configuration, Fenn Mill.
- SEM microscopy was used to establish lamination window with great interlayer contact at microscopic level.
- Evaluated the interplay of ASL thickness, processibility and mechanical strength.
- EEA with minimal target thickness of ASL (~300 µm) was fabricated via slot-die coating.

Fanny May Mill

Laminated EEA
ASL / ASL / AFL / GDC





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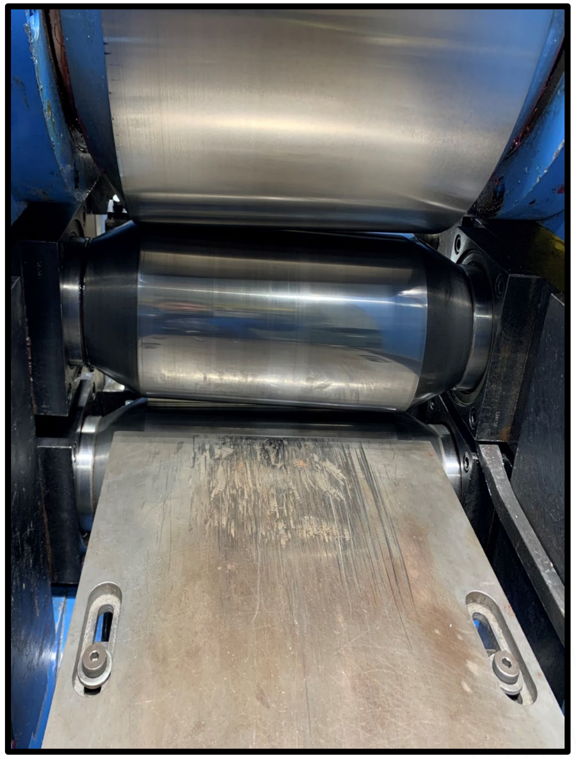
Fenn Mill at ORNL together with an Oven used in Calendering Experiments

- The individual, coated layers were laminated on the Fenn Mill to form EEA.
- Prepared EEA with minimal ASL thickness (300 μm per layer).
- Identified lamination window and process parameters (temperature, gap, speed, sequence).



Sample preparation: 1.5” W x 4” L samples of ASL - ASL/AFL/GDC, ASL-ASL-AFL/GDC and ASL-ASL-AFL-GDC.
7” x 7” ASL – ASL/AFL/GDC
90^{o}C, 110^{o}C, 130^{o}C temperature equilibration of 5 minutes.

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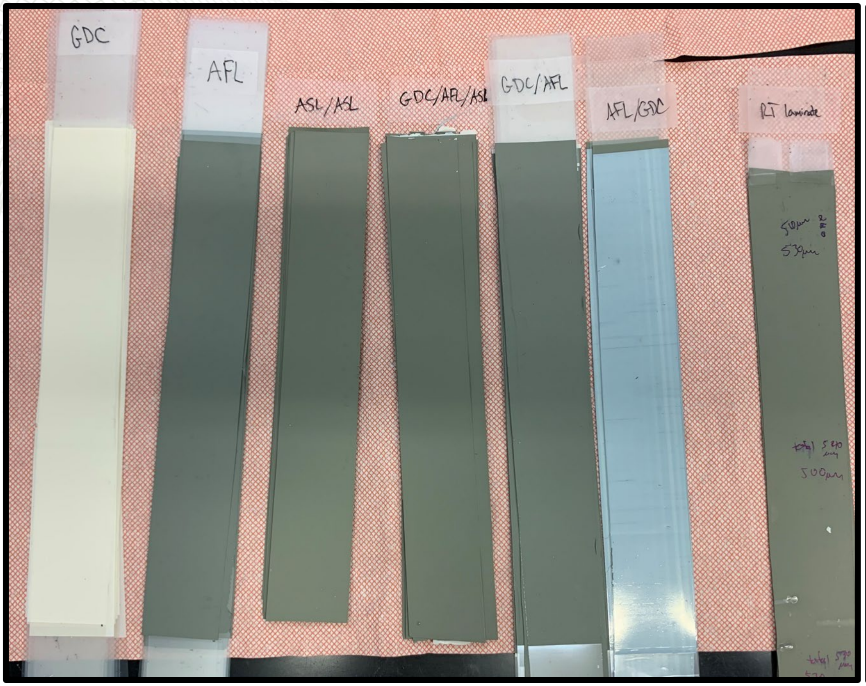
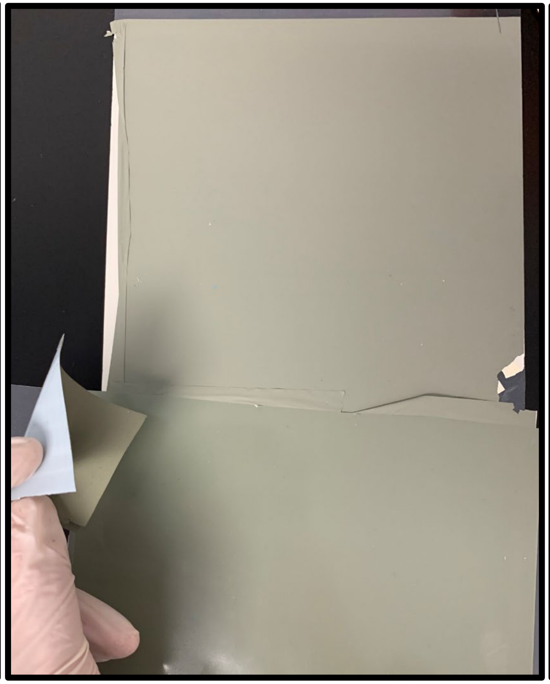
Excellent lamination at 90^{o}C, resulting in 6 to 9% reduction





- Lamination of single- and multi- layer coatings results in great contact on macroscopic level within the established processing window.
- SEM was used to evaluate microscopic contact between layers and confirm integrity of the EEA.

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1-Pass ASL-ASL/AFL/GDC Calendering at 130°C, 110°C and 90°C

Temp. (° C)	Set Gap (um)	Speed (FPM)	Initial thickness (um)	Final thickness (um)	Reduction thickness (um)	Compressed percentage (%)
1 pass: ASL-ASL/AFL/GDC, total EEA width: 410 um						
130	431.8	20.0	410	378	32.0	7.8%
1 pass: ASL-ASL/AFL/GDC, total EEA width: 410 um						
		25.0	410	382	28.0	6.8%
130	381	20.0	410	379	31.0	7.6%
		15.0	410	369	41.0	10.0%
		2.0	410	378	32.0	7.8%
1 pass: ASL-ASL/AFL/GDC, total EEA width: 410 um						
		25.0	410	377	33.0	8.0%
130	355.6	20.0	410	358	52.0	12.7%
		15.0	410	358	52.0	12.7%
1 pass: ASL-ASL/AFL/GDC, total EEA width: 410 um						
		25.0	410	390	20.0	4.9%
130	330.2	20.0	410	381	29.0	7.1%
		15.0	410	360	50.0	12.2%
1 pass: ASL-ASL/AFL/GDC, total EEA width: 442 um						
		25.0	442	390	52.0	11.8%
110	381	20.0	442	385	57.0	12.9%
		15.0	442	384	58.0	13.1%
1 pass: ASL-ASL/AFL/GDC, total EEA width: 442 um						
		25.0	442	392	50.0	11.3%
110	355.6	20.0	442	382	60.0	13.6%
		15.0	442	380	62.0	14.0%
1 pass: ASL-ASL/AFL/GDC, total EEA width: 442 um						
		25.0	442	395	47.0	10.6%
110	330.2	20.0	442	385	57.0	12.9%
		15.0	442	385	57.0	12.9%
1 pass: ASL-ASL/AFL/GDC, total EEA width: 455 um						
		25.0	455	414	41.0	9.0%
90	381	20.0	455	409	46.0	10.1%
		15.0	455	405	50.0	11.0%
1 pass: ASL-ASL/AFL/GDC, total EEA width: 410 um						
		25.0	410	369	41	10.0%
90	330.2	20.0	410	374	36	8.8%
		15.0	410	363	47	11.5%

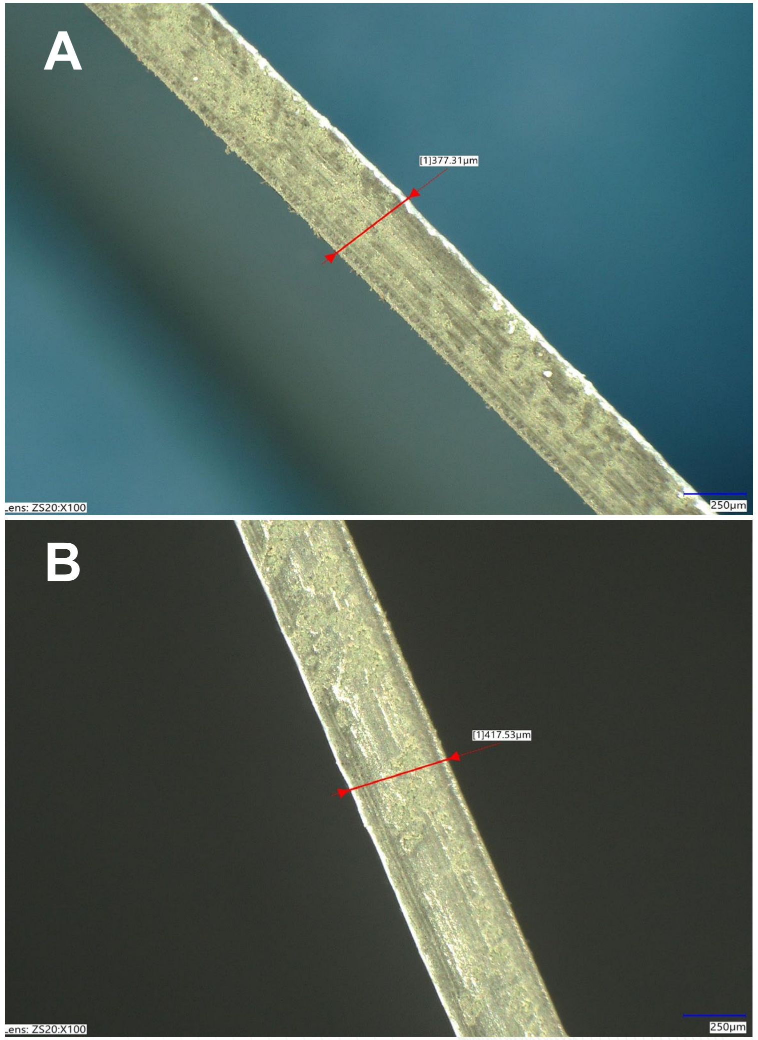

Figure above shows optical micrograph at 100X magnification of EEA sample prepared through 1 pass calendering at a gap of 381 um. The ASL green sheet was laminated onto a triple-layer coated green sheet of ASL/AFL/GDC at A) at 130 °C and an initial thickness of 410 um, resulting in 6.8% reduction of thickness to 382 um and B) at 90 °C and an initial thickness of 455 um, resulting in 9.0% reduction to 414 um.

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1-Pass ASL-ASL-AFL/GDC & 2-Pass ASL-ASL-AFL-GDC Calendering at 90^{o}C

Temp. (^{o} C)	Set Gap (um)	Speed (FPM)	Initial thickness (um)	Final thickness (um)	Reduction thickness (um)	Compressed percentage (%)	Comments
1 pass: ASL-ASL-AFL/GDC, total EEA width 277 um							
90	330.2	25.0	277	252	25.0	9.0%	Sticks
		20.0	277	245	32.0	11.6%	Sticks
1 pass: ASL-ASL-AFL/GDC, total EEA width 277 um							
90	304.8	25.0	277	240	37.0	13.4%	Sticks
		20.0	277	235	42.0	15.2%	Sticks
		15.0	277	233	44.0	15.9%	Sticks
2 pass: ASL-ASL-AFL-GDC total EEA width 380 um							
90	457.2	25.0	380	343	37.0	9.7%	Sticks
		20.0	380	339	41.0	10.8%	Sticks
		15.0	380	340	40.0	10.5%	Sticks
2 pass: ASL-ASL-AFL-GDC total EEA width 380 um							
90	431.8	25.0	380	344	36.0	9.5%	Sticks
		20.0	380	337	43.0	11.3%	Sticks
		15.0	380	337	43.0	11.3%	Sticks
2 pass: ASL-ASL-AFL-GDC total EEA width 367 um							
90	431.8	25.0	367	322	45.0	12.3%	Sticks
		20.0	367	322	45.0	12.3%	Sticks
		15.0	367	315	52.0	14.2%	Sticks
2 pass: ASL-ASL-AFL-GDC total EEA width 367 um							
90	406.4	25.0	367	322	45.0	12.3%	Sticks
		20.0	367	321	46.0	12.5%	Sticks
		15.0	367	315	52.0	14.2%	Sticks
2 pass: ASL-ASL-AFL-GDC total EEA width 367 um							
90	381	25.0	367	316	51.0	13.9%	Sticks
		20.0	367	311	56.0	15.3%	Sticks
		15.0	367	309	58.0	15.8%	Sticks
		3.0	367	311	56.0	15.3%	Sticks

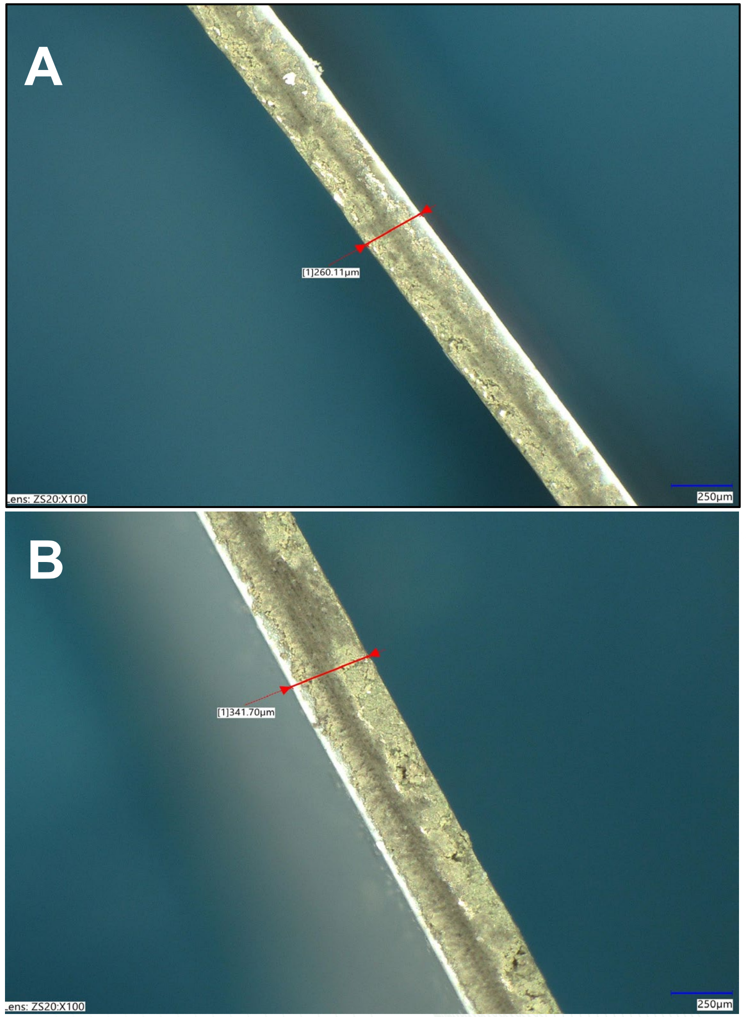

Figure above shows optical micrograph at 100X magnification of EEA sample prepared at 90 ^{o} C through A) 1 pass calendering with a set gap of 330.2 um, where 2 individual ASL green sheets were laminated onto a double-layer coated green sheet of AFL/GDC resulting in a 9.0% reduction in thickness to 252 um from 277 um. B) 2 pass calendering, first of the GDC onto the AFL followed by addition of 2 ASL layers onto the assembly, laminating at a set gap of 457.2 um. The initial thickness of 380 um was compressed by 9.7% down to 343 um.

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Summary

- Completed fabrication of EEA via slot die coating and lamination, establishing stable processing windows for both processes.
- Fabricated EEA with varying ASL thickness down to minimal thickness of 300 μm.
- Lamination window was confirmed at microscopic level via SEM imaging.
- High throughput fabrication can be achieved under strict control of processing parameters.

Future work in FY24

- Sintering the multilayer EEA and comparing the properties with those from traditional batch process
- Assembling SOFC button cells and evaluating electrochemical performance

Acknowledgements

- Office of Fossil Energy and Carbon Management
- Project manager: John Homer
- ORNL Team
- Redox Power Systems Team

Thank You

Any Questions?