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U.S. DEPARTMENT OF ENERGY
Hydrogen and Fuel Cell Technologies Office

R2R: Roll-to-Roll Consortium

Scott Mauger
National Renewable Energy Laboratory
May 8, 2024
WBS – 12.3.0.501/13.2.0.501

DOE Hydrogen Program
2024 Annual Merit Review and Peer Evaluation Meeting

MNF-BIL001

NREL Transforming ENERGY
Argonne NATIONAL LABORATORY
BERKELEY LAB
OAK RIDGE National Laboratory
Sandia National Laboratories

Project Goal

Mission: Advance efficient, high-throughput, and high-quality manufacturing methods and processes for clean hydrogen technologies to accelerate domestic manufacturing and reduce the capital cost of durable and high-performing systems

Goals:

1. **Collaborate with industry** to develop scalable, affordable manufacturing processes and enable technology transfer
2. **Understand material and process behavior** to advance methods to increase energy efficiency, reduce material usage, reduce hazardous waste, and reduce equipment cost in manufacturing processes
3. **Enable high-throughput processes** (while maintaining performance) that industrial partners can *demonstrate*
4. **Validate manufacturing cost model assumptions** for/toward near-term capacity targets

Overview

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Timeline and Budget

- Project Start Date: 10/1/2023
- FY23 DOE Funding: $500k (FY24 forward funding)
- FY24 Planned DOE: $6.625M
- Total DOE Funds Received to Date: $7.125M

Barriers

- Lack of high-volume MEA processes
- Low levels of quality control
- Cost

National Lab Consortium Team

NREL Transforming ENERGY · Argonne NATIONAL LABORATORY · BERKELEY LAB Bringing Science Solutions to the World · OAK RIDGE National Laboratory · Sandia National Laboratories

Subcontract

- Strategic Analysis, Inc – fuel cell manufacturing cost analysis
- Univ. of New Mexico – coating process modeling

Overview

- 5-year project (10/1/2023 – 9/30/28)
- $50M anticipated funding (50/50 FC/WE)
- Core lab R&D
  - 6 task areas
  - Initial focus on PEM FC and WE
- Industry Support
  - CRADAs
  - FOA support

Potential Impact: Scale Up is Challenging!

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Battery Plant Scrap Rates Can Hit 90% At Ramp Up, But The Situation Is Improving

It can take a company years to make up for the massive costs of starting up a battery plant.

BY JIM MOTAVALLI PUBLISHED: FEB 5, 2024

https://www.autoweek.com/news/a46628833/early-production-battery-plant-scrap-rates/

"It can be a three- or four-year journey to get to profitability."

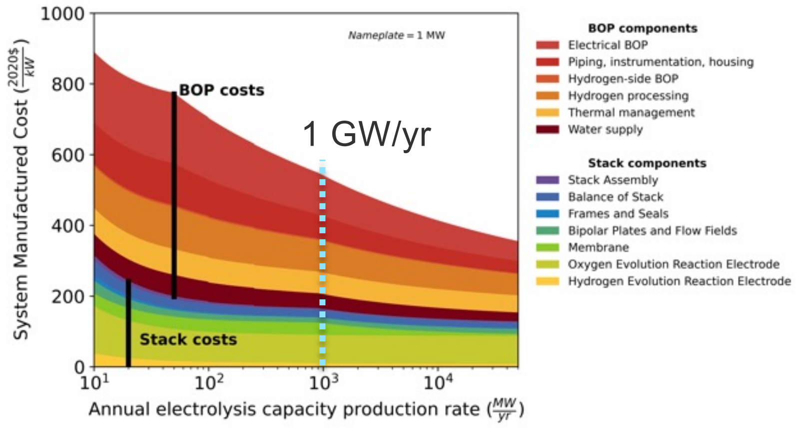
"…reducing scrap rates by just one percentage point can mean tens of millions in added profit annually."

Challenges in Scale Up

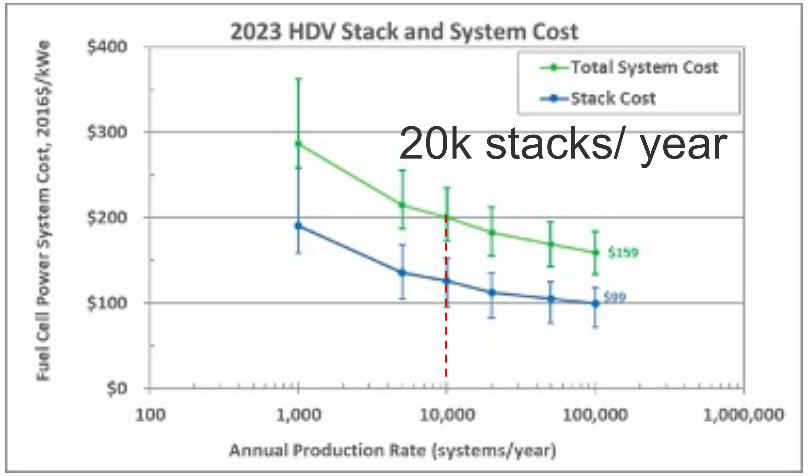
- Critical materials data gap
- Process complexity
- Inadequate models
- Perceived barriers
- Unknown unknowns

Adapted from Miller, et al., Risk Minimization in Scale-Up of Biomass and Waste Carbon Upgrading Processes. *ACS Sustainable Chem. Eng.* **2024**, *12* (2), 666–679. https://doi.org/10.1021/acssuschemeng.3c06231.

Potential Impact



Badgett, et al., Updated Manufactured Cost Analysis for Proton Exchange Membrane Water Electrolyzers. 2024. Golden, CO: National Renewable Energy Laboratory. NREL/TP-6A20-87625. https://www.nrel.gov/docs/fy24osti/87625.pdf.



James, et al., *Mass Production Cost Estimation of Direct H2 PEM Fuel Cell Systems for Transportation Applications: 2023 Update on Heavy and Medium-Duty Vehicles*; 2023.

- Increase in manufacturing scale needed to meet FOA 2922 and Hydrogen Shot targets
- Roll-to-roll manufacturing key to meeting production rate targets of FOA 2922:
  - PEMWE: > GW/yr
  - HD FC: 20k stacks/yr
- Performance and durability targets also critical

- Relevant DOE Goals
  - Strengthen U.S. Manufacturing
    - R&D focused on advancing manufacturing process science for PEMWE and FC
  - Provide pathways to private sector uptake
    - CRADA projects with industry

Approach – Where we fit in

Make better materials	*Make better components*	*Better make components*	*Manufacture materials/stacks*
HydroGEN Advanced Water Splitting Materials; ElectroCat Electrocatalysis Consortium	H2NEW Hydrogen from Next-generation Electrolyzers of Water, U.S. Department of Energy; M2FCT Million Mile Fuel Cell Truck	Roll to Roll Consortium, U.S. Department of Energy, Hydrogen and Fuel Cell Technologies Office	Industry

Proof-of-Concept | **Information Oriented** | **Profit Oriented**

Information Oriented R&D

- Rigorous evaluation and resolution of process risks
- Determine key rates and parameters
- Address key perceived barriers

Approach – Synergizing Multiple Efforts

R2R brings together multiple project efforts into a single consortium

Roll-to-Roll Advanced Materials Manufacturing Collaboration (TA007, 2021)
- AMO-funded with HFTO contribution
- Scaled synthesis processes
- Machine learning
- Process modeling and development

M2FCT/H2NEW
- Concentrated inks
- R2R coating processes
- Drying

MEA Manufacturing R&D (TA001)
- QC tool and method development
- Spatial diagnostic studies
- Modeling of defect impacts

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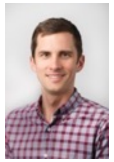
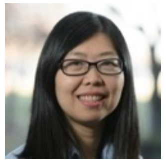
Approach: Consortium Organization

Director
Scott Mauger (NREL)



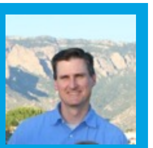


Deputy Directors
Yuepeng Zhang (ANL)
Alexey Serov (ORNL)

M2FCT/H2NEW Liaison
Debbie Myers (NREL)

Materials Scale-Up Science
Jessica Macholz (ANL)

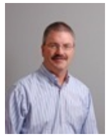
MEA Fabrication Process Science
Nelson Bell (SNL)



Quality Control
Peter Rupnowski (NREL)

Characterization
Vince Battaglia (LBNL)

Advanced Computing
Randy Schunk (SNL)



Technoeconomic Analysis
Alex Badgett (NREL)

Approach – Go/No-go Milestones

Description	Criteria	Date
Using R2R coating, fabricate catalyst layers with improved uniformity compared to baselines established earlier in FY24 and measure MEA initial performance. Uniformity improvements may include reductions in cross-web or down-web loading uniformity, concentration of discrete defects, or ionomer-catalyst ratio. Use resulting process data to conduct technoeconomic analysis of unit costs for comparison against earlier established baseline and consortium objectives.	• Coating Length: at least 5 m • Uniformity quantified from at least 10 locations • MEA testing performed on at least 3 MEAs	9/30/2024

Manufactured MEA cost will be metric used to track consortium progress

Quarterly progress measures shown in backup slides

Approach: Materials Scale-Up Science

Research Objectives

- Improve mixing, heat transfer, and reactant distribution to achieve large-scale, continuous/batch synthesis of complex materials
- Improve process accuracy, reproducibility, and efficiency
- Scale-up of next-generation materials developed through M2FCT, H2NEW, and other HFTO-supported programs/projects
- Increase scientific understanding of large-scale synthesis

Year 1 Activities

- Scaling BNL M2FCT catalyst
- Establish relationships between synthesis process conditions, product properties, product quality and performance

Approach: MEA Fabrication Process Science

Research Objectives

- Inks/Dispersions
  - Link materials properties to ink properties
  - Improved stability
  - Faster mixing
- Coatings
  - Establish process windows
  - Link ink properties to coating quality and microstructure
  - Coating processes for next-gen components or improved processes
  - Predictive process maps for R2R methods
- Drying
  - Improvements in drying efficiency
  - Link drying process to layer microstructure/properties
  - Develop drying models to predict and dictate structure and for process optimization

Approach: MEA Fabrication Process Science

Year 1 Activities

- Inks
  - Conduct experiments and build models to link materials properties to ink properties
  - Formulations to improve ink stability
  - Investigation of mixing methods
- Coating Processes
  - Establish baseline coating processes
  - Process window studies for R2R catalyst layer coating
  - Link ink properties to coating defects
  - Investigate impacts of different coating substrates (liner, membrane, GDL)
- Drying
  - Develop methods, equipment, and models for evaluation of advanced drying methods
  - Study links between drying and layer structure and defects

Approach: Quality Control

Research Objectives

- Quality monitoring tools
  - Evaluate and demonstrate applicability of various high throughput in-line QC methods
  - Develop novel and customized QC techniques
  - Integrate QC tools with AI/ML for process improvements and control
- Impact of manufacturing variations on device behavior
  - Correlate manufacturing variations with performance and durability using in-situ spatial diagnostics cell hardware
  - Conduct device modeling to predict impact of defects and help define QC method requirements
  - Testing quality and uniformity of commercial materials available on the market to determine QC needs and to understand correlation between material variations and ultimate device performance limitations

Year 1 Activities

- Develop and validate new techniques, testbeds, and measurement capabilities
- Performance and durability testing of fuel cell and electrolyzer MEAs with known heterogeneities and/or defects
- Develop models to support QC method development and assess impact of defects on performance and simulate defect-induced deformations

Approach: Characterization

Research Objectives

- Validation and development of characterization methods and platforms for the manufacturing environment
- MEA performance and durability validation testing to link process to performance
- Utilize national lab capabilities (TEM, SAXS, etc.) to study impacts of process conditions on morphology, composition, etc.

Year 1 Activities

- Evaluation of particle sizing methods for concentrated catalyst inks
- Particle size/shape analysis to link particle structure to ink properties
- Develop in situ drying analysis tool for confocal microscopy and x-ray radiography to characterize drying films

Approach: Advanced Computing

Research Objectives

- Multiscale multiphysics models for advanced processes
- Develop real-time data streaming and AI data analytics to achieve dynamic interaction with in-situ sensors and in-line metrology
- Develop multifidelity ML models using all collected computational and in-line and off-line experimental data
- Enable automated process control, monitoring, and optimization

Year 1 Activities

- Modeling of coating and drying processes for non-Newtonian fluids and different drying modes
- CFD modeling of reactors to improve products
- Develop database schema and ML framework for data fusion from all resources for ML enabled correlation of materials-process-performance relationships

Approach: Technoeconomic Analysis

Research Objectives

- Develop more granular modeling frameworks for specific manufacturing approaches and technologies, identifying cost and yield characteristics of different approaches
- Leverage experimental R&D across the consortium to inform model inputs, key cost metrics, and opportunities to reduce costs while increasing yields and performance

Year 1 Activities

- Granular analysis of existing and near-term manufacturing processes and materials for at-scale manufacturing
- TEA focusing on process optimization and emissions estimates of existing materials and equipment

Approach: CRADA Projects with Industry

- DOE-cost shared projects between R2R labs and industry
- Details:
  - Up to $8M ($1.5M project max)
  - 2-year projects
  - 50% cost share
- ADD URL Here
- Webinar: May
- Proposals due June 16, 2024

- Technologies: PEM, AEM, liquid alkaline
- Topics include:
  - Scaled synthesis processes
  - R2R process modeling and development
  - QC method development and/or validation or
  - ML/AI
  - Characterization (electrochemical, X-ray, microscopy)

Approach: Safety Planning and Culture

- Safe operating procedure are in place for the different work activities
- Researchers are required to complete targeted safety trainings to ensure safe completion of work (e.g. platinum group metal safety, nanomaterials)
- Safety reviews are conducted prior to beginning new work or significant change in scope
- This project was not required to submit a safety plan to the Hydrogen Safety Panel

Approach:
Established Industrial Advisory Board

- Andrew Motz, Nel Hydrogen
- Nathan DeMario, Plug
- Michelle Ostraat, Pajarito Powders
- Andy Smeltz, De Nora USA
- Narendra Deshpande, General Motors
- Marc Gurau, Chemours
- Deepak Shukla, Ionomr Innovations
- Chris Kerscher, Association for Roll-to-Roll Converters

Board will advise consortium on research activities and priorities

Accomplishments and Progress: Held Kickoff Workshop with Industry

- August 2023 workshop introduced consortium to US hydrogen industry
- Approximately 80 industrial attendees from 31 companies
- Selected industry-identified needs:
  - Studies related to product quality, yield, reproducibility
  - Tolerance specifications/impacts of variation
  - Ink stability
  - Process modeling (reactors, coating, drying)
  - Process – structure links/predictive modeling
  - QC methods

Accomplishments and Progress: Machine Learning Data Collection for Consortium-Wide Experiments

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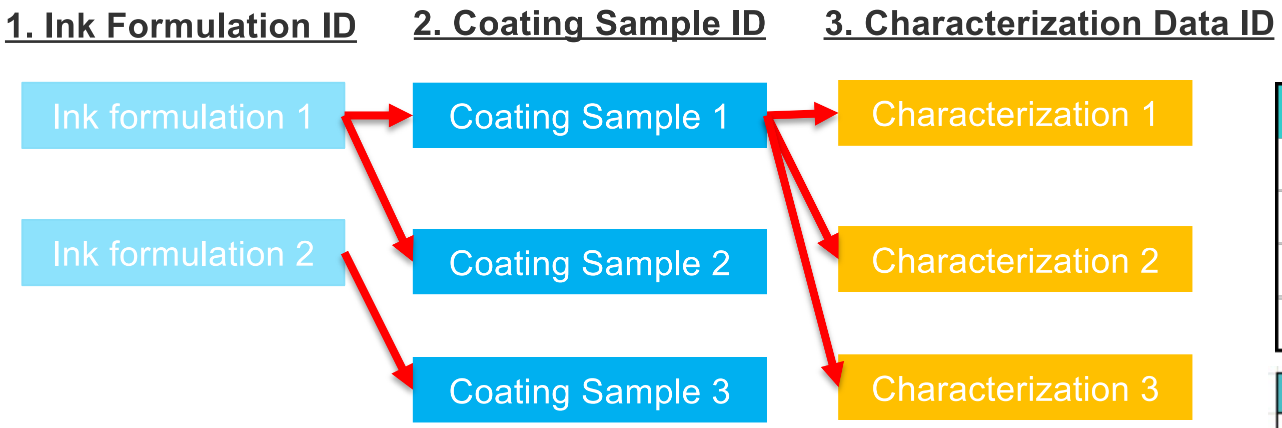
- Consortium-wide data collection needed to train multi-fidelity machine learning (ML) models
- Standardizing data reporting and formatting is necessary to ensure information accuracy and compatibility
- Designed data reporting standard for process and characterization steps



- Established unique identifiers to link ink formulations, coating samples, and characterization measurements
- Implemented dropdown options, data validation rules, and pre-defined property fields to **standardize formatting and reduce data entry errors**

Example: ink metadata

Box 1. Generate a unique ID for your metadata		
	Today's Date	2/27/2024
	Lab Identifier	ANL
Make new ID	Your Initials	AL
	Sample ID	/_240213_ANL_AL_580

Box 2. Enter metadata		
Catalyst material	IrO2	Select a dropdown option.
Support material	Vulc	Select a dropdown option.
Catalyst loading on support (wt%)	[illegible]	Enter a value.
Ionomer solution name	Nafion D2020CS	Select a dropdown option.
Catalyst + support (wt%)	25	Enter a value.
Ionomer solution (wt%)	25	Enter a value.
Alcohol solvent type	1-propanol	Select a dropdown option.
Added water (wt%)	25	Enter a value.
Added alcohol (wt%)	25	Enter a value.

Accomplishments and Progress: Initiated Process Development of M2FCT Catalyst

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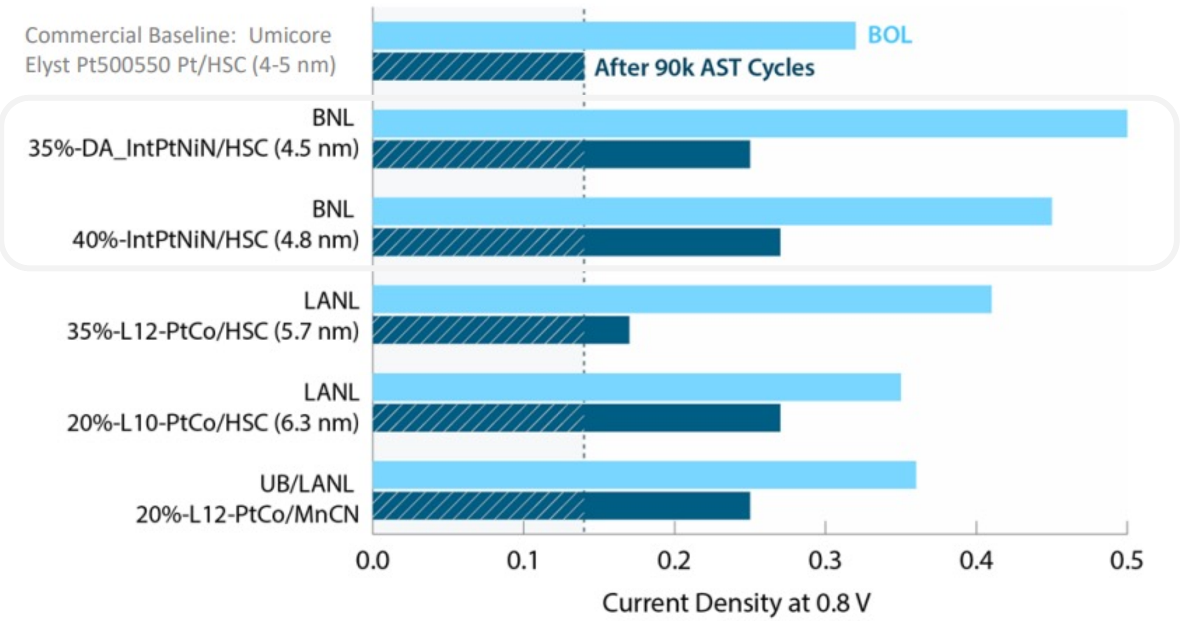
- To successfully transition new materials to market, research, development, and demonstration are required to gain a better understanding of the materials synthesis process and to develop scalable approaches that improve manufacturability
- R2R leadership under guidance from DOE determined a catalyst from M2FCT to scale up and coordinated with Brookhaven National Laboratory (BNL) on the synthesis details

M2FCT CATALYST PERFORMANCE



Performance measured at Beginning of Life (BOL) and after 90,000 AST cycles

Image: https://www.hydrogen.energy.gov/docs/hydrogenprogramlibraries/pdfs/review22/plenary7_papageorgopoulos_2022_o.pdf

- **BNL's synthesis procedure was sent to R2R labs to prepare for scale-up:**
  - **Year 1: Duplicate material with similar properties through scalable synthetic approaches**
  - **Year 2: Scale to larger amounts**

Accomplishments and Progress: Catalyst Synthesis Using Batch Reactor

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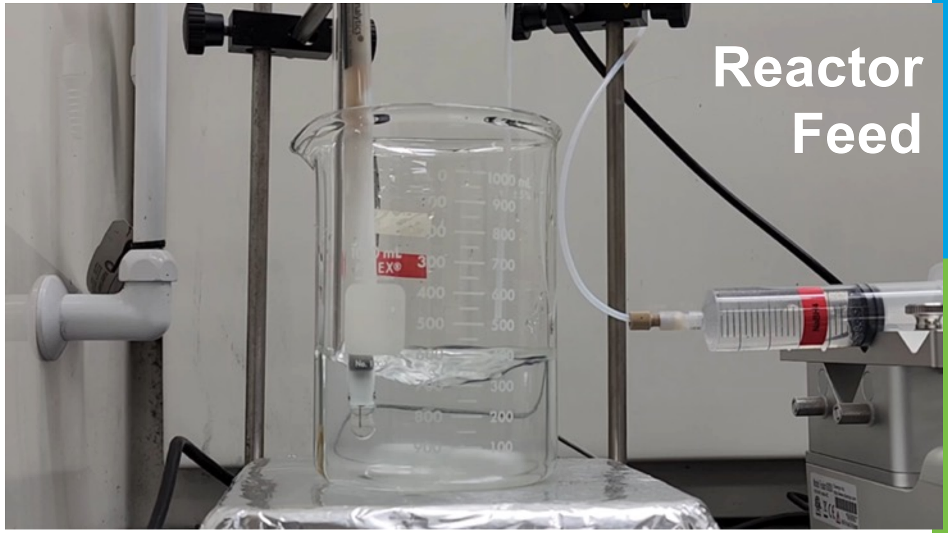
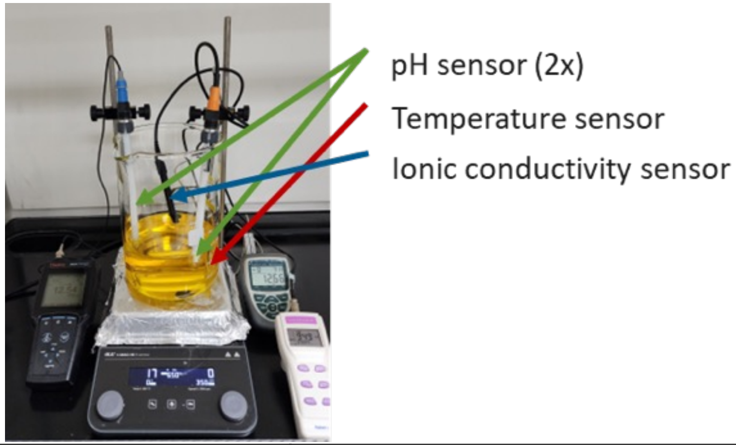
- Model experiments conducted with Ag-nanoparticles
- Impact of RPM, impeller type, and presence of capping agent (citric acid) was studied
- Obtained data will be fed to CFD modelling of a reactor



Reactor with Sensors to Monitor Conditions in Real-Time



Experimental Parameter	Information Obtained	Reactor Improvement
pH	Gradient of pH, rate of pH equilibrium, local pH	Rate of acid/base addition, position of addition, stirring rate
Temperature	Gradient of T, rate of T equilibrium	Stirring rate, solution volume, reactor shape
Ionic Conductivity (IC)	Gradient of IC, rate of IC equilibrium, local IC	Rate of reagents addition, position of addition, stirring rate
Average Nanoparticle Size	Rate of particle growth, particle size distribution	Stirrer geometry, stirring rate, reactor shape, position of reagents addition

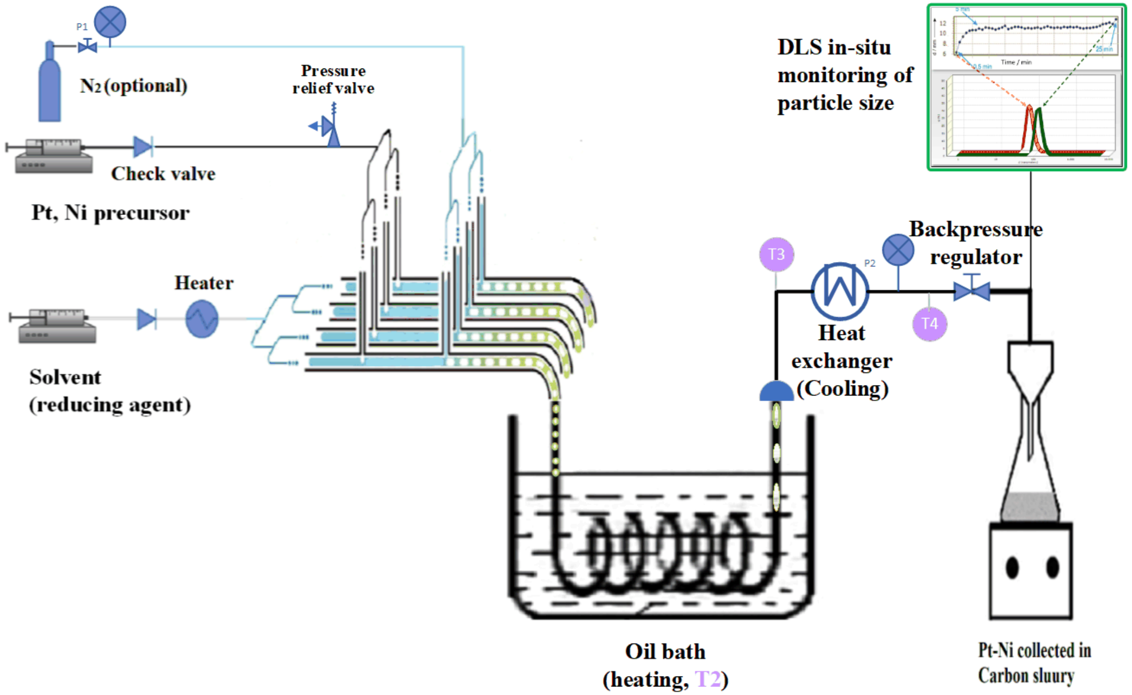
Sample	Particle size (nm) *Ag, 2 peaks*	Particle size (nm) *Ag, all peaks*	RPM	$NaBH_4$ (g/50mL)	Stirrer
Ag-1A	56.3	51.7	100	2	IKAFLON 80
Ag-2A	49.5	56.5	200	2	IKAFLON 80
Ag-3A	47.1	53.5	300	2	IKAFLON 80
Ag-4A	74.4	126.1	100	1	IKAFLON 80
Ag-5A	54.7	65.3	200	1	IKAFLON 80
Ag-6A	51.3	49.3	300	1	IKAFLON 80

- **In general, with increase of RPM, particle size decreases (baffled system)**
- **Higher concentration of $NaBH_4$ resulted in smaller particles**
- **Precise reagent addition and sensors reading is required to obtain structure-to-parameters correlation**

Accomplishments and Progress: Catalyst Synthesis Using Flow Reactor

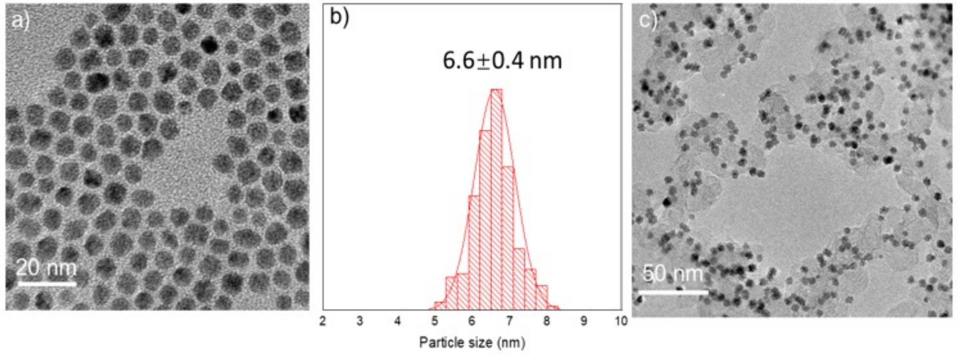
- A parallel continuous flow reactor has been designed and fabricated
- In-line monitoring will allow us to explore the influence of synthesis conditions on the physical and chemical characteristics of the catalyst

Scalable, Continuous Flow Reactor



Schematic of a parallel continuous flow reactor with in-line monitoring for scale-up

Catalyst Synthesized using a Scalable, Continuous Flow Reactor (Prior Work)

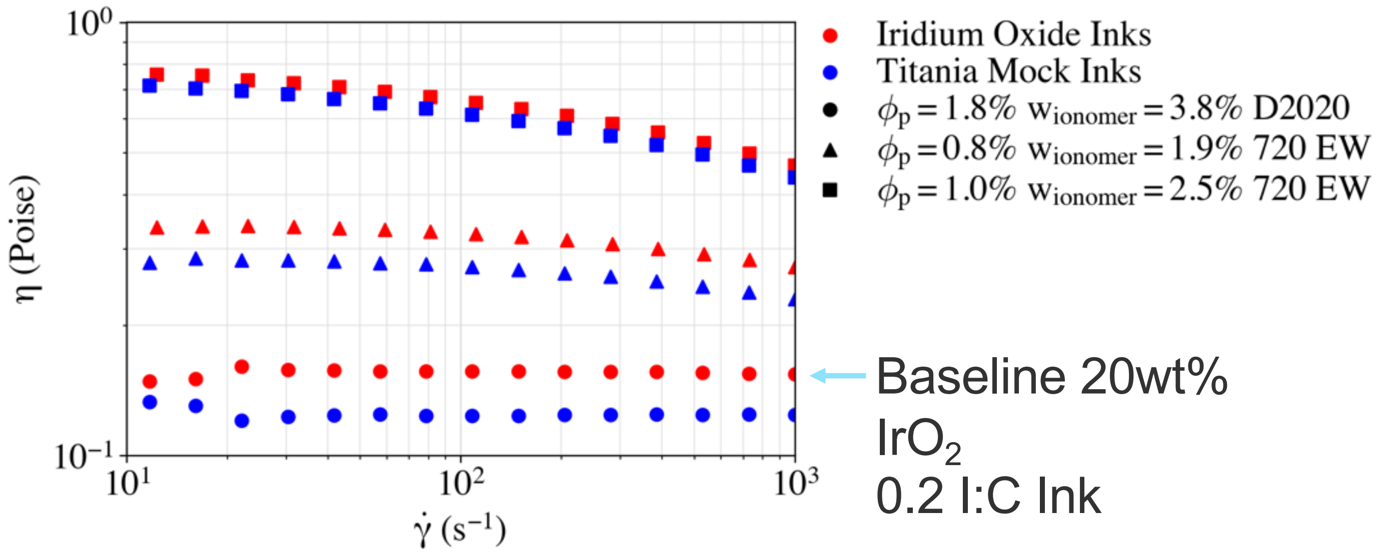


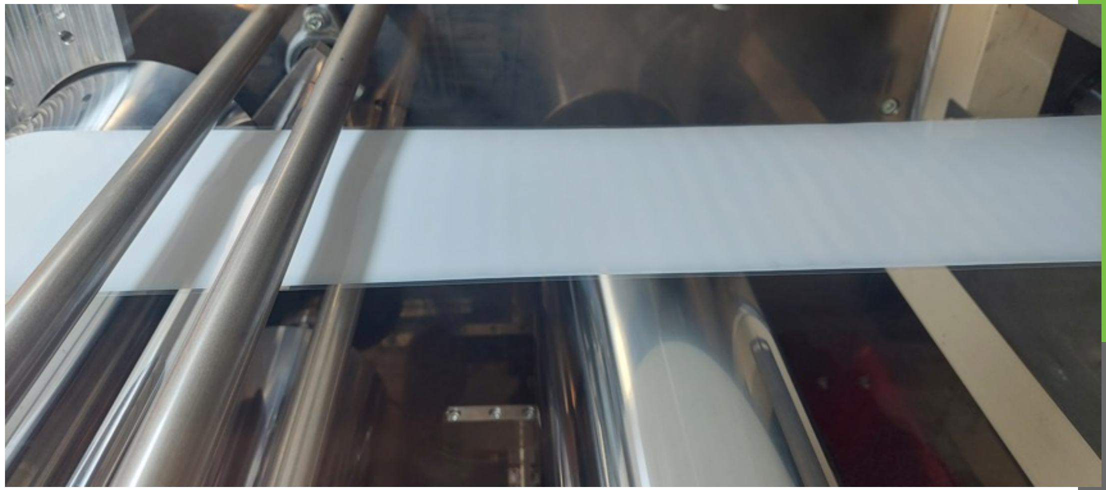
Transmission electron microscopy (TEM) images of a PtCoNi catalyst synthesized using a continuous flow reactor before (*left*) and after (*right*) carbon load

- **Synthesize PtNi in a newly developed parallel continuous flow reactor (PCFR) and nitride PtNi using a scalable fluidized bed reactor**
- **The PCFR procedure will be scaled by increasing precursor flow rate and concentration along with coil tubing dimensions and quantity**
- **Process automation will be investigated via *in-situ* monitoring (e.g., particle size using dynamic light scatting, DLS) and AI/ML**

Accomplishments and Progress: Mock Inks

- Mock inks are needed to mitigate cost barriers for comprehensive R2R coating studies
- Mock and real inks were prepared with similar catalyst volume fractions (ϕ_p) and ionomer mass fraction ($w_{ionomer}$) to target similar properties
- Directly addresses industry-stated need from Kickoff Workshop

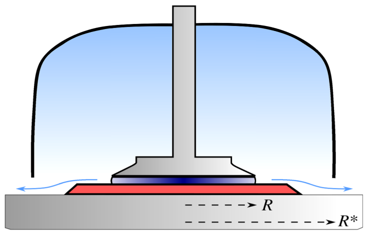




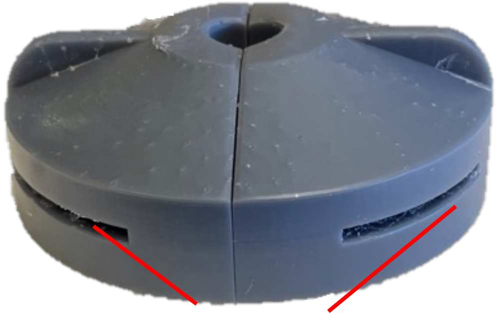
- **Mock and iridium oxide inks possessed similar rheologies**
- **Similar defects were observed between iridium oxide and mock ink coating trials**
- **Ongoing work is exploring known causes of these defects and further characterization**

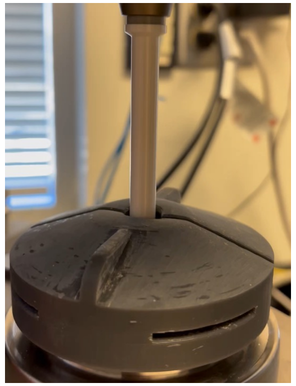
Accomplishments and Progress: Development of Tools for Drying Studies

Measurement of Rheology During Drying



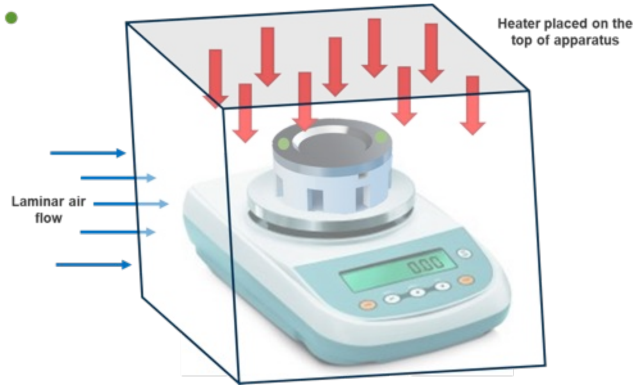
[1] Lehéricey, P and Divoux, T. *et al. J. Rheo.* (2021) 427 – 436





- Zero normal force measurement allows for gap adjustment as sample volume decreases
- Conducting testing for optimized cap design and rheology protocol.
- Output data:
  - Rheology as a function of solids volume fraction
  - Constitutive equation for the fluid during the drying process

Drying Apparatus



- Drying rate measurements provide diffusion coefficients for ink compositions.
- New instrument configuration desired to incorporate greater testing features.
  - Addition and control of laminar flow rate and air temperature
  - Changeable heating source: NIR, microwave, or combination
  - Thermal measurement of sample pan and drying chamber

Accomplishments and Progress: Response to Reviews

- This project was not reviewed last year

Collaboration and Coordination

HFTO-funded

- Work closely to ensure use of relevant materials, metrics, methods, and protocols
- Develop scale-up solutions for materials and structures developed in other HFTO-funded projects to demonstrate pathway to manufacturing
- Leverage M2FCT/H2NEW expertise and methodologies for advanced characterization and testing

Industry

- IAB will be used to guide research R2R research activities
- CRADA projects for collaborative R&D

BIL Initiatives

- Providing primary resource consumption data to ANL for BIL project life cycle analysis efforts

DEIA/Community Benefits Activities

- Consortium CBP is under development
- CRADA projects planned to include CBPs

DEIA/CB Activities

- SNL subcontract with U. New Mexico (MSI) for coating process modeling
- Workforce Development (FY24 current and planned):
  - Post docs: 10
  - Graduate students: 5
  - Interns: 3

Remaining Challenges and Barriers

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- Advances in fuel cell and electrolyzer manufacturing needed to meet Hydrogen Shot and other HFTO goals
- Cost modeling assumes increases in scale can be accomplished without losses in performance and durability, but this must be demonstrated and validated
- Need to develop and validate formulations and processes for high-yield coating processes
- Need for full-web inspection tools

Proposed Future Work

Materials Scale-Up
- Link process to product quality
- Scale M2FCT catalyst

MEA Fabrication Process Science
- Inks
  - link materials properties to ink properties
  - Improve ink stability
  - Investigation of mixing methods
- Coating Processes
  - Establish baseline coating processes
  - Process window studies for R2R catalyst layer coating
  - Link ink properties to coating uniformity/defects
  - Investigate impacts of different coating substrates
- Drying
  - Evaluation of advanced drying methods
  - Link drying to layer structure and defects

Quality Control
- Develop and validate new techniques/methods
- Link heterogeneities/defects to performance/durability
- Models to support QC method development and assess impact of variation/defects

Characterization
- Evaluation of particle sizing methods
- Particle size/shape analysis
- Develop in situ drying analysis tools

Advanced Computing
- Modeling of coating and drying processes for non-Newtonian fluids and different drying modes
- ML enabled correlation of materials-process-performance relationships
- Reactor CFD

Technoeconomic Analysis
- Extend analysis framework to emerging and innovative manufacturing processes and materials
- Develop iterative TEA process with experimental and AI/ML tasks to map cost impacts of experimental work in the consortium and guide R&D strategies
- TEA focusing on process optimization and emissions estimates of existing materials and equipment

CRADA Projects

Any proposed future work is subject to change based on funding levels.

Acknowledgements

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SNL: Nelson S. Bell, Mariah Gallegos, Erik Linde, Rekha Rao, P. Randall Schunk

HFTO: McKenzie Hubert, Eric White, Mike Ulsh, Greg Kleen, Julie Fornaciari, Dave Peterson, Dimitrios Papageorgopolous

Summary

- New consortium addressing process science and manufacturing for fuel cells and electrolyzers
- Mission: Advance efficient, high-throughput, and high-quality manufacturing
- Research focused on enabling manufacturing at HFTO target rates (FC: 20k stacks/yr, WE: >GWs/yr)
- Combination of Lab-led R&D and CRADA projects with industry