# Membrane Electrode Assembly Manufacturing Automation Technology for the Electrochemical Compression of Hydrogen

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Project ID: h2006

# Overview

## Timeline and Budget

- Project start date: 05/01/18
- Original project end date: 11/01/19
- Extended end date: 8/1/20
- Total project budget: $371,240
  - Total partner share: $150,000
  - Total federal share: $150,000
  - Total partner in-kind: $71,240
  - Total funds spent*: $250,517

* As of 4/30/20

## Barrier

- Reliability and Costs of Gaseous Hydrogen Compression (Delivery B)

## Partners

- HyET
- National Renewable Energy Laboratory (NREL)
- Lawrence Berkeley National Laboratory (LBNL) (associated membrane project with HyET)

# Relevance

- Relevance (from $H_2$@Scale CRADA call):
  - Develop materials, processing techniques, and/or innovative designs for components used in hydrogen equipment and compressors, to enhance durability
  - Design **novel manufacturing approaches** and technologies for compressors and pipeline fatigue life and durability
- Objectives:
  - Full rheological understanding of **optimum ink formulation** for roll-to-roll coating method for catalyst layers, followed by electrochemical hydrogen compression (EHC) relevant tests
  - Development of **real-time optical/areal quality inspection** techniques for membrane electrode assembly (MEA) materials, with a focus on the membrane
  - Compare **EHC performance** of roll-to-roll (R2R) and manually manufactured MEAs (HyET)
  - Design and specification of an **EHC MEA manufacturing line** (HyET with NREL)

# Approach

- Project leverages NREL in-line MEA inspection and electrode scaling capabilities and expertise developed for fuel cell and electrolysis materials, and apply to EHC
- NREL is developing quality control (QC) techniques and inks/coating process understanding and transferring knowledge to HyET
- HyET is performing in situ testing of coated materials in their hardware/system
- Project outcomes will inform manufacturing line specification

# Accomplishments and Progress

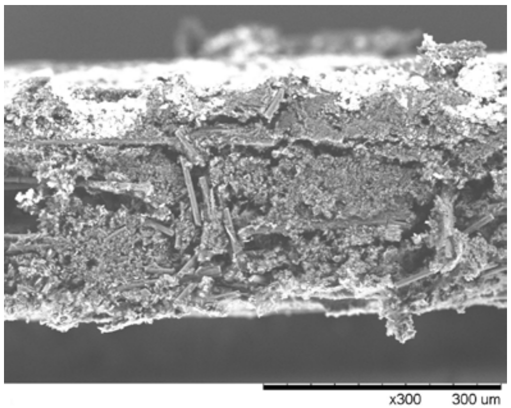
**Review: Coated electrodes using R2R slot-die**

- Coating onto two different GDLs
  - 18 cm wide, both substrates
  - Three target loadings
  - Turrax homogenizer, then stirred overnight to degas
  - Web-speed 1 m/min
  - Oven 1: 80 C, oven 2: 90 C
- Used cross-sectional scanning electron microscopy (SEM) to study electrode structure
- Validated electrode loading and variability via XRF
- Electrodes coated at all conditions and provided to HyET for in situ testing
- *Expanded our understanding of formulating inks for and coating onto porous substrates*

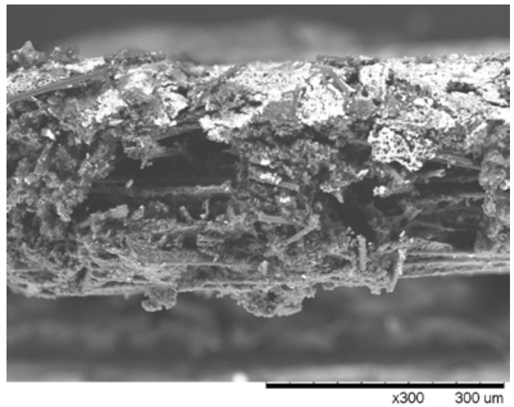
***Presented in 2019***

# Accomplishments and Progress

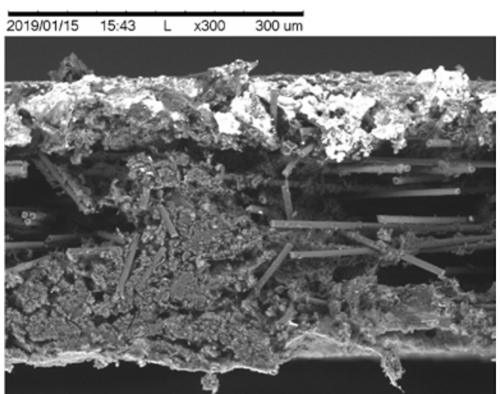
## Cross-section SEM: Verified minimal ink penetration (on both substrates) with NREL-developed inks and coating processes (compared here to HyET screen printed baseline)

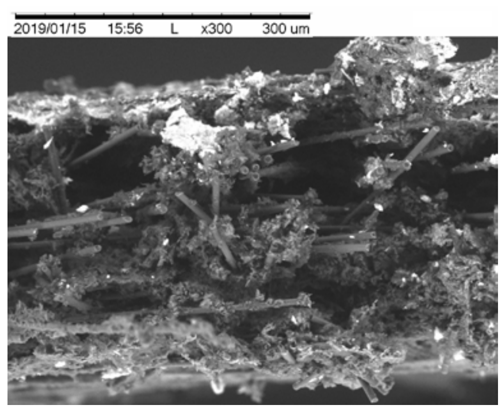



Screen printed HyET baseline




Spray coated

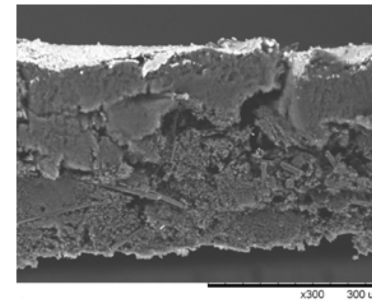


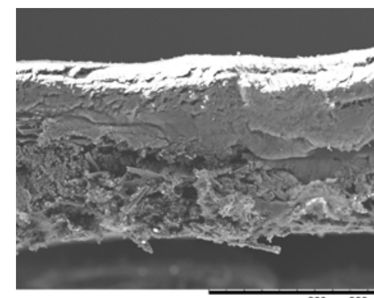

R2R slot die coated: 1X (left) and 2X (right) catalyst loading

GDL A

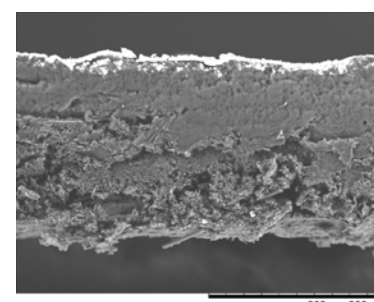
GDL B




Screen printed HyET baseline



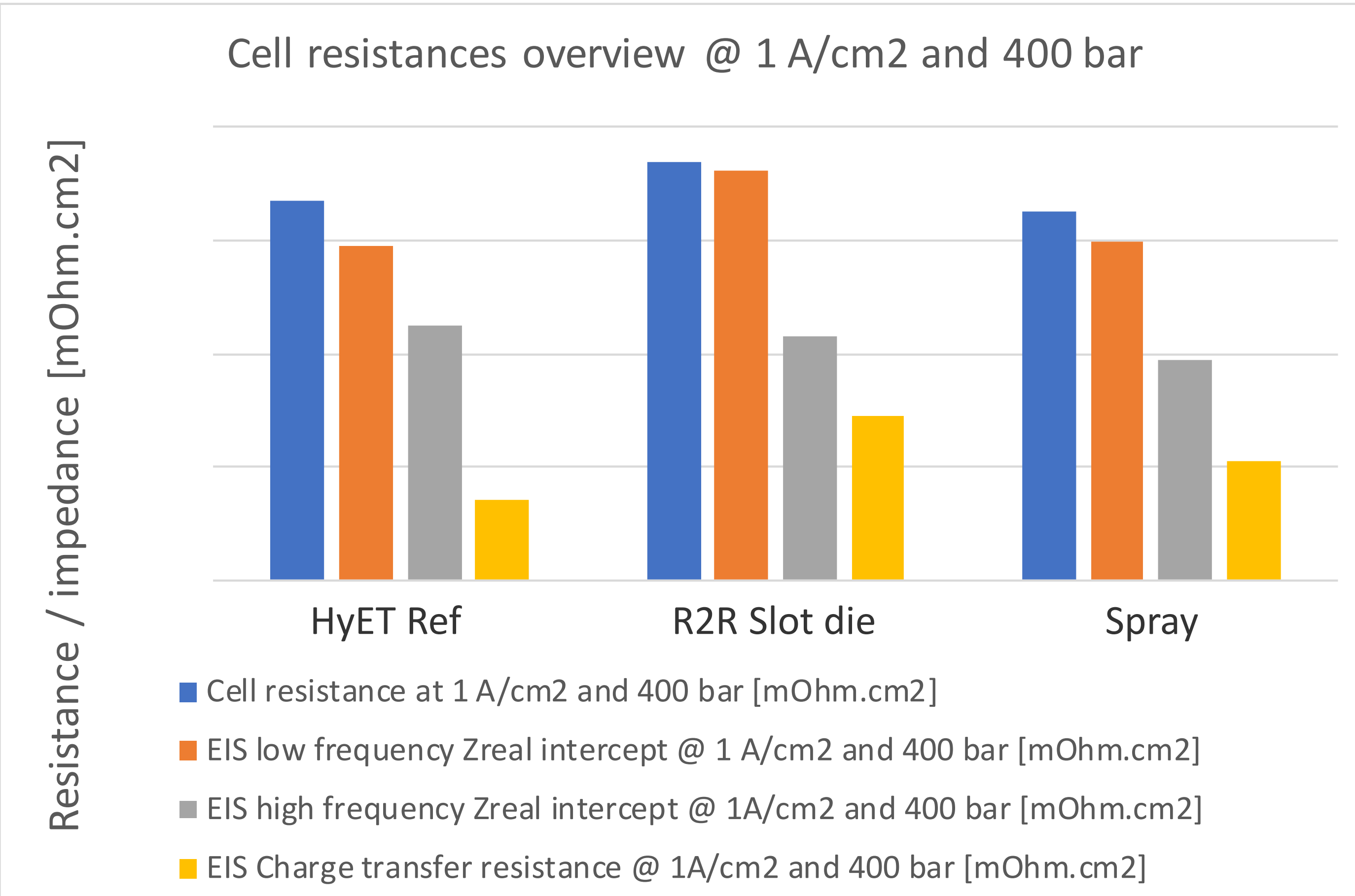

Spray coated




Rod coated

# Accomplishments and Progress

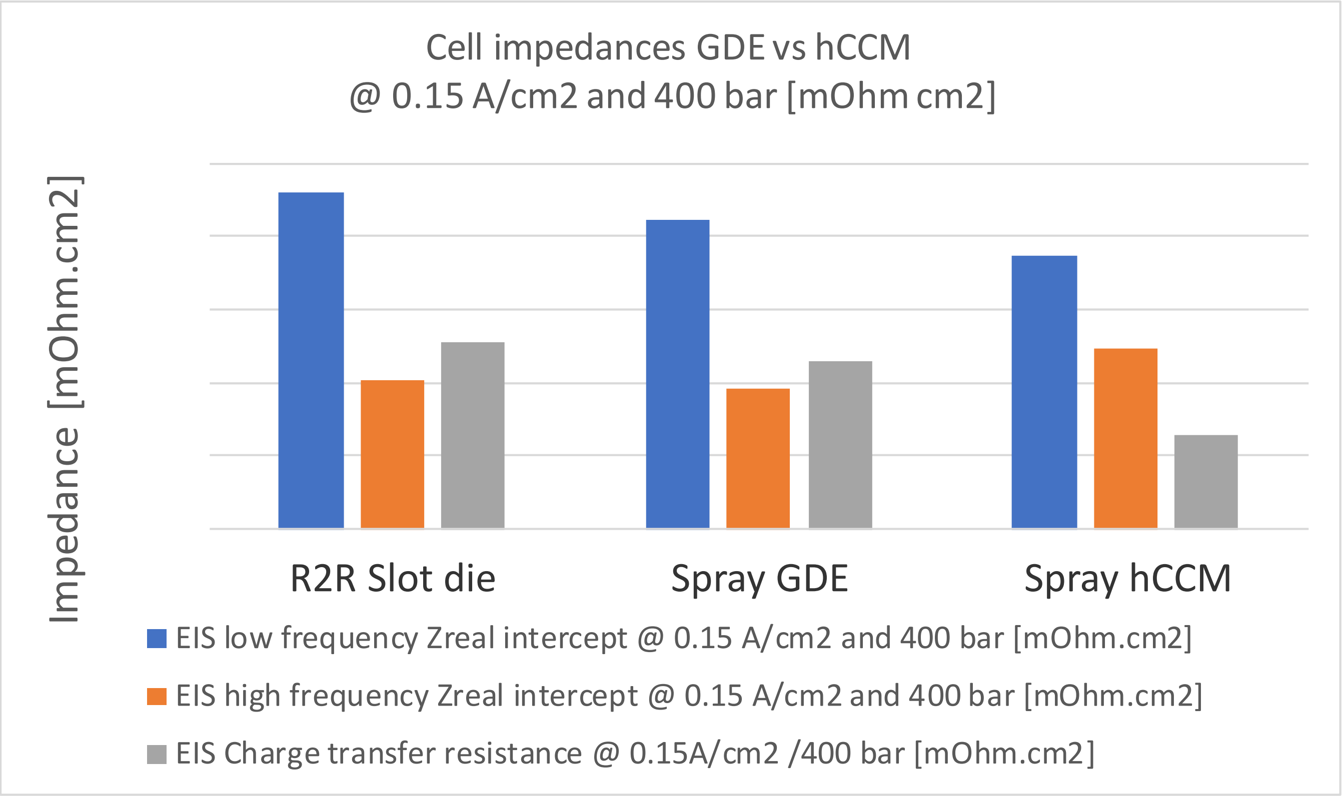
## Performed in situ EHC testing of NREL anodes (HyET): HyET reference vs. Spray vs. Slot die GDEs




- Sprayed GDE has comparable resistances to HyET reference process
- R2R slot die GDE had higher resistances

# Accomplishments and Progress

## Performed in situ EHC testing of NREL anodes (HyET): R2R GDE vs. Sprayed GDE vs. Sprayed half-CCM (hCCM)




- hCCM has significantly lower charge transfer resistance than GDEs
- hCCM had the lowest EIS low frequency intercept
- High frequency intercepts were comparable

# Accomplishments and Progress

## Performed in situ EHC testing of NREL anodes (HyET): Sprayed hCCMs at different loadings

| Cell | Pt loading | Resistance |
|---|---|---|
| Spray hCCM | ~1 | ~0.67 |
| Spray hCCM | ~0.25 | ~1 |
| Spray hCCM | ~0.5 | ~1 |
| Reference GDE | 1 | 1 |

*Resistance at 400 bar in galvanostatic compression test*

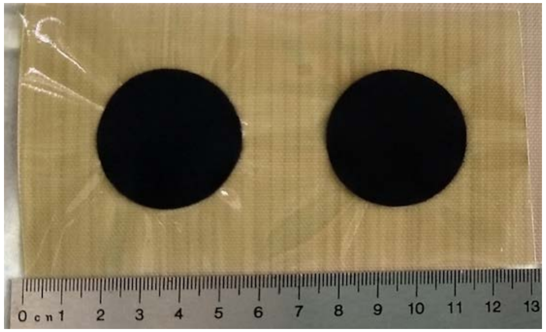
- All hCCMs outperformed the reference GDE
- Spray hCCMs had comparable performance to reference: cost reduction opportunity
- Spray hCCM at same loading had significantly less resistance
  - Improved membrane-electrode interface for hCCM
  - Ink recipe change

# Accomplishments and Progress

**Explored patterned electrode coating to reduce catalyst waste**

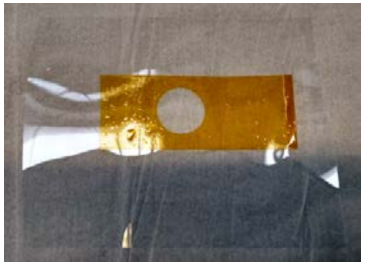
- Spray coating using mask

| Ink formulation (10 g basis) | | | |
|---|---|---|---|
| Catalyst [g] | DI water [g] | n-propanol [g] | Ionomer [g] |
| 0.4 | 6.678 | 1.856 | 1.066 |

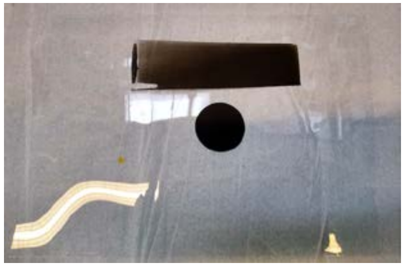


Replicate pattern spray coated electrodes on HyET membrane

- Rod coating using tape mask
  - 5 wt% catalyst, 1:1 nPA:DIW, 1:1 I:C ink



Kapton tape mask applied to PET



Coated circle after removal of Kapton tape mask

Coated electrode on HyET membrane

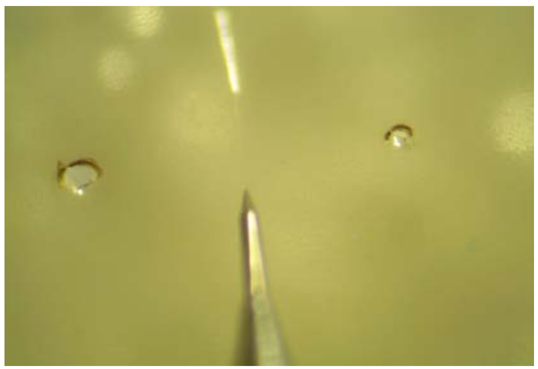
*Both methods showed promise for further development, e.g. using R2R gravure printing*

# Accomplishments and Progress

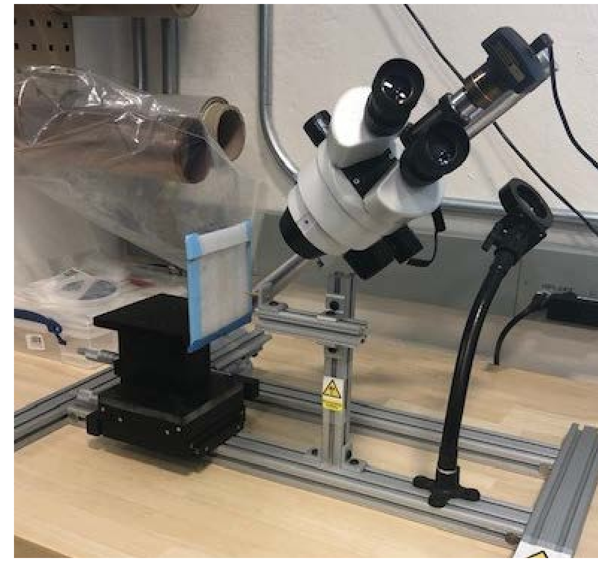
## Fabricated membranes with pinholes to evaluate impact on in situ performance

- Methodology

In-process visualization of tool creating pinholes, 30X magnification



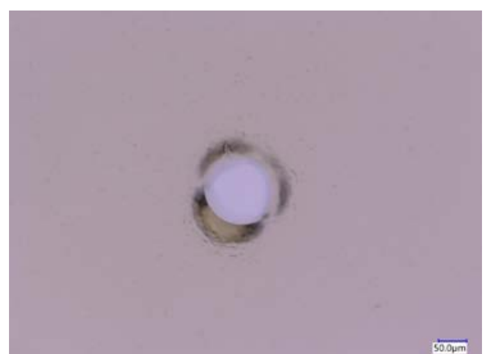
Fabrication setup with optical microscope, 3-axis control stage, and sample support



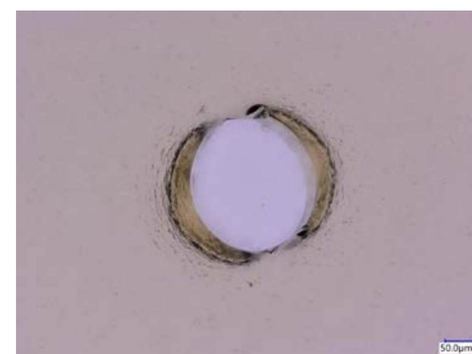
- Results

Digital microscope, 400X

Pinhole using 25 µm tool



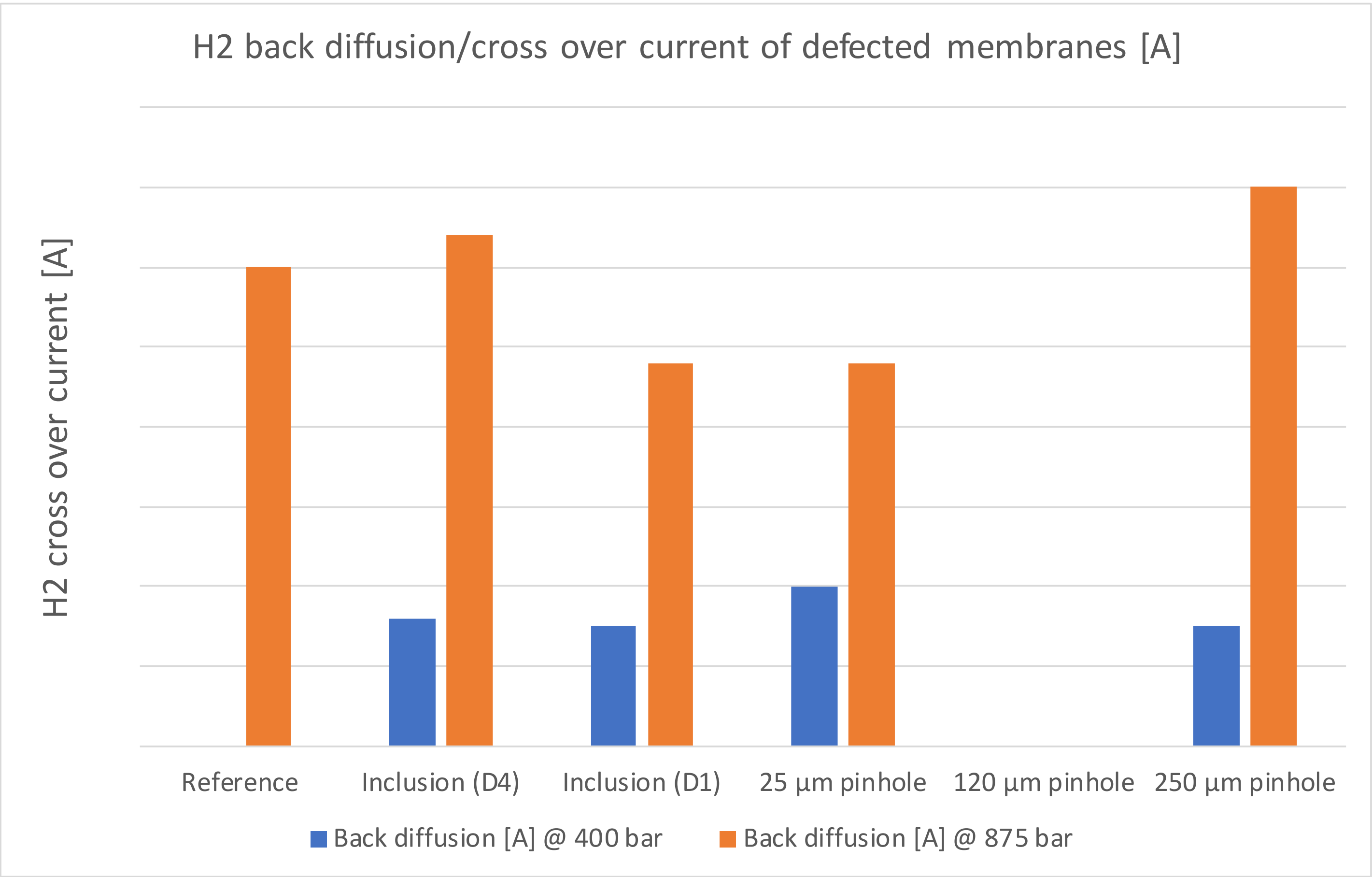
Pinhole using 120 µm tool



Pinhole using 250 µm tool

# Accomplishments and Progress

## Performed in situ EHC testing of membranes with pinholes to evaluate impact on in situ performance (HyET)




- Membranes with pinholes and included particles did not appear to have a significant impact on H2 crossover current density
- *MEA hot lamination may partially mitigate defect impact*

# Accomplishments and Progress

**Developed MEA manufacturing pathway scenarios (NREL & HyET)**

- Assess sub-assembly process scenarios, starting with cases with most confidence (e.g. state of the art or known industry use), and building to more efficient cases that require further development
  - Included sourcing vs. internal manufacturing, sub-assembly structures, and cell assembly
- Assist HyET in evaluating sourcing options, vertical integration, and design and flexibility of R2R manufacturing equipment

# Accomplishments and Progress: Responses to Previous Year Reviewers' Comments

- This project was presented as a poster, but not reviewed last year

# Collaboration and Coordination

- Industry partner: HyET
  - Defines objectives
  - Defines materials and structures
  - Provides information about applicable manufacturing processes and techniques
  - Performs in situ cell testing of NREL-produced cell materials
  - Defines major parameters for MEA manufacturing line
- National lab partner: NREL
  - Explores and develops relevant inspection techniques for MEA materials
  - Formulates and characterizes inks for scalable electrode processing
  - Explores applicable coating processes
  - Provides methods and coated materials to HyET
  - Assists in the specification of MEA manufacturing line
- Additional partner: LBNL
  - LBNL has membrane-focused project with HyET; NREL and LBNL will coordinate on research as applicable

# Challenges and Barriers

- Formulating inks for scalable processes
- Identifying scalable process methods and procedures for high-volume electrode production
- Understanding manufacturing pathways, including sourcing and integrated paths, and equipment options for MEAs
- Exploring gravure printing of electrodes to reduce catalyst waste

# Proposed Future Work
## (Based on remaining already received project funding)

- Optimize ink formulation for uniform coating of GDEs, based on the prior small-scale and R2R experiments.
- Optimize ink formulation and determine suitable substrate for small-scale patterned electrode fabrication on decal, based on the prior patterning work.
- Develop inks and R2R gravure printing for patterned R2R anode fabrication on a decal.
- Fabricate full CCMs.
- Explore ink development and coating of high porosity substrates.
- *HyET to perform in situ testing of NREL samples.*

# Technology Transfer Activities

- Under the CRADA, applicable technology will be transferred to the industry partner
- NREL will assist HyET in the implementation of project outcomes

# Summary

- Approach: leverage NREL manufacturing R&D capabilities developed for fuel cells to similar EHC materials and processes
- Relevance: develop materials, processes, techniques and manufacturing approaches for electrochemical hydrogen compression MEAs
- Accomplishments
  - Cross-section SEM to evaluate ink penetration
  - In situ testing (at HyET) of NREL electrodes
  - Patterned electrode coating
  - Membrane defect fabrication and in situ testing (at HyET)
  - Manufacturing pathway scenarios
- Future Work
  - Finalize GDE inks and coating
  - Small-scale and R2R decal coating, with CCM fabrication
  - Gravure printing development
  - Exploration of coating on very porous substrates

# Acknowledgements

HyET

- Martijn Mulder, Industry Partner PI
- Albert Bos, CTO
- Melissa Bosch, R&D engineer
- Rick Jansen of Lorkeers, R&D engineer
- Alexis Dubois, Project Manager

NREL

- Peter Rupnowski, QC Development sub-lead
  - Brian Green
- Scott Mauger, MEA Scaling sub-lead
  - Jason Pfeilsticker
  - Sunil Khandavalli

LBNL

- Gao Liu, Sister-project collaborator

# Thank You

**www.nrel.gov**

Publication Number

**NREL is a national laboratory of the U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy, operated by the Alliance for Sustainable Energy, LLC.**

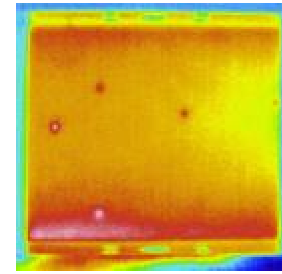
# Technical Back-Up Slides

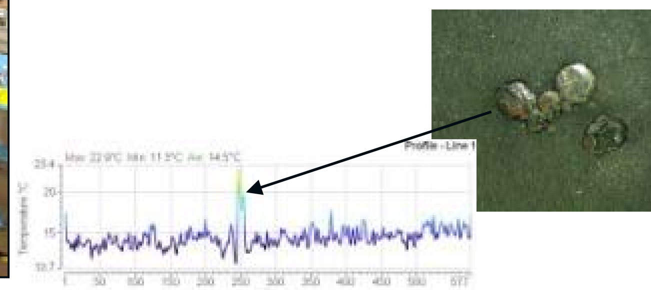
# NREL Capabilities: In-line Inspection

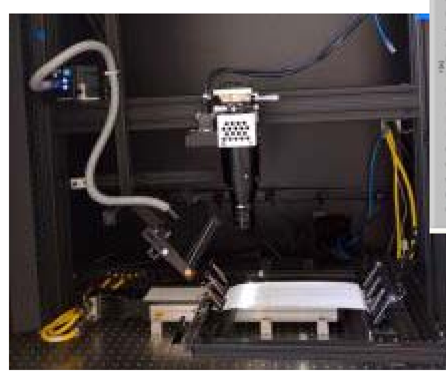
**Development of real-time, in-line quality control diagnostics to support the continued scale-up of MEA components for low-cost production**

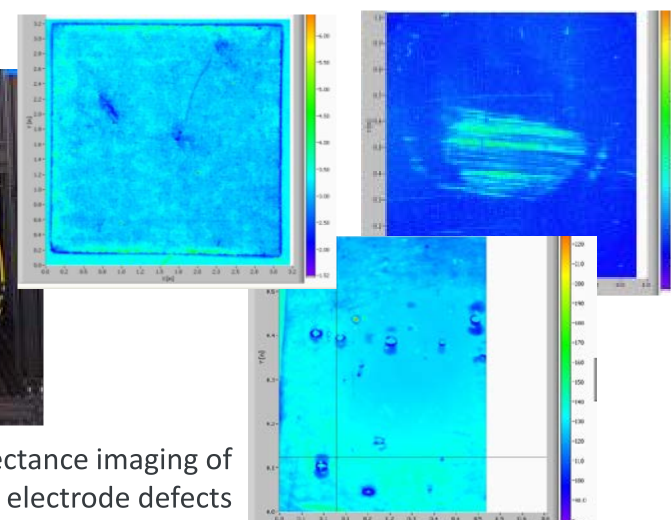
- Material portfolio: Fuel cell, battery, and electrolyzer materials
- Membrane defect imaging and thickness mapping
- Electrode and GDL uniformity
- MEA shorting and gas crossover
- Property measurement, e.g. porosity
- Optical and IR diagnostic platforms
- Non-destructive, 100% inspection

Infrared imaging of electrode and MEA defects









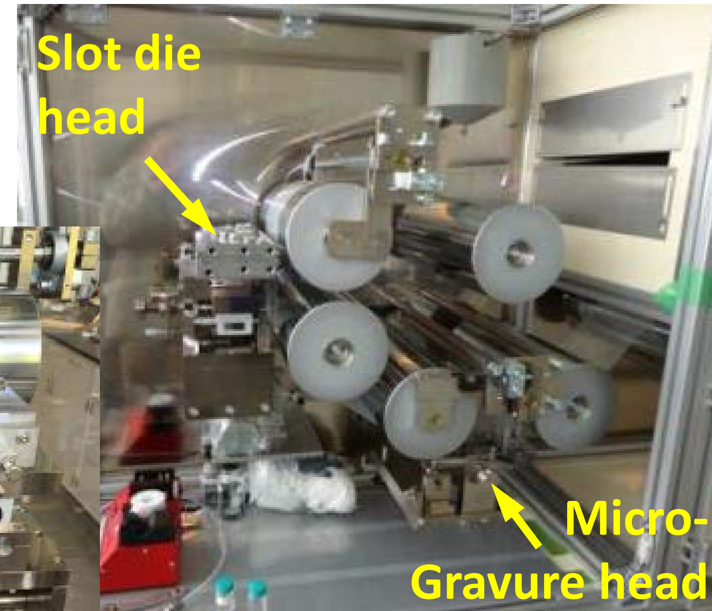
Optical reflectance imaging of membrane and electrode defects

# NREL Capabilities: MEA Scaling

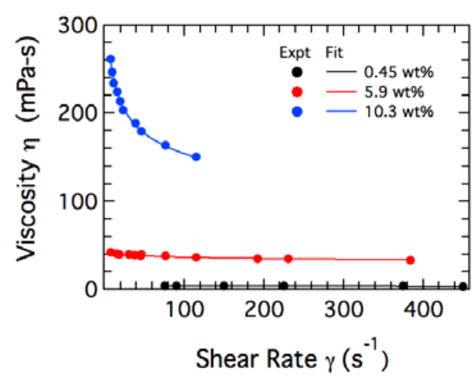
**Understanding how parameters of scalable, high-volume processes effect MEA morphology, uniformity, and performance**

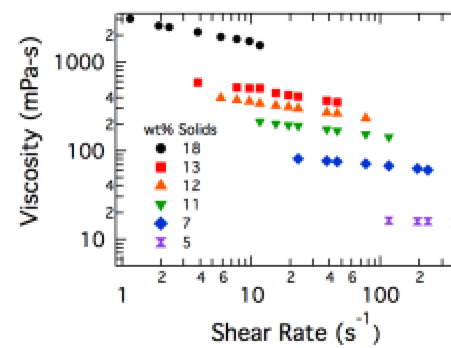
- Roll-to-roll coating station
  - Slot/knife coating head
  - Micro-gravure coating head
  - 50-300 mm coating width
  - 4 independently controllable drying sections
  - Coating speeds from 0.2-10 m/min
- Applicable to various thin-film technology applications
- Formulation, rheology, coating, drying studies

Slot-die coating




Multi-head coating enclosure




Viscometry of electrode inks

# Project Task Details

- **Task 1: Inks and Coating Process Development (NREL)**
  NREL will bring to bear ink characterization tools, including rheology, dynamic light scattering, and zeta potential, and electrode fabrication equipment across several scales including a roll-to-roll coating line to evaluate and perform initial optimization of inks and coatings for EHC electrodes. NREL will perform parametric studies to understand the impact of formulation and process variables on the thickness and uniformity of electrode layers. NREL will provide electrode sheet materials to HyET for assembly into cells and in situ testing at HyET's facility.

- **Task 2: Quality Inspection Development (NREL)**
  NREL will bring to bear multiple test-beds for the development of real-time quality inspection techniques for electrodes, membranes, and MEA subassemblies for HyET's EHC MEA materials. NREL's activity focuses on areal inspection techniques, i.e. techniques that utilize optical or infrared imaging to provide the potential for 100% inspection of MEA material webs (rather than point measurements). These techniques enable detection of small discrete defects in MEA materials as well as determination of overall film or layer uniformity. If appropriate, NREL can utilize its industrial-style web-line to validate techniques with sheet or roll materials.

# Project Task Details

- **Task 3: EHC testing of R2R vs manual MEA based EHC single cell & stack (HyET)**

  HyET will manufacture the MEAs and perform EHC cell testing which will include standard diagnostics (as used for PEMFC) like shorting tests, IV-curve recording, (Electrochemical Impedance Spectroscopy) EIS and cyclic voltammetry (CV analysis), hydrogen (H2) cross-over testing, H2 pumping at ambient pressure and finally compression testing to maximum pressure. Duration testing to evaluate the performance stability will conclude the membrane evaluation test program. Performance of the manual vs automated manufactured MEAs will be compared.

- **Task 4: Specification of a manufacturing Line (HyET+NREL)**

  Using its experience with R2R equipment and fabrication of MEA materials, and based on its work and results in Tasks 1 and 2, NREL will assist HyET in the design and specification of a manufacturing line for the high volume production of its MEAs. NREL will specifically provide inputs relative to electrode coating and quality inspection devices and integration. US suppliers of required production line equipment will be identified.