Influence of Coating Method on the Performance of Roll-to-Roll Coated PEM Fuel Cell Catalyst Layers

Scott Mauger
20th ISCST Symposium
September 21, 2020
Proton Exchange Membrane Fuel Cells

- Light duty vehicles (LDV), heavy duty trucking, trains, marine transportation, stationary power
- High efficiency (>2x internal combustion)
- Zero GHG emissions
- Long-range EV (LDV - 300+ miles/tank)
- Short refueling time (LDV - 5 min)

https://www.nrel.gov/hydrogen/
https://nikolamotor.com/one
Catalyst layer microstructure impacts fuel cell performance

Unknown how R2R coating methods impact catalyst layer structure
Experimental

Slot Die Coating

Gravure Coating

- Web Speed: 1 m/min
- Oven Temp: 80 °C
- Substrate: diffusion media


Fuel Cell Performance

At 100% RH
- No differences in kinetics
- Slot die better at high current densities

At 40% RH
- Large difference in kinetics
- Slot die better at all current densities
Electrochemical Surface Area

- \( \text{Pt Util.} = \frac{ECSA(X \%RH)}{ECSA(100\%RH)} \)
- At high RH, little effect of coating method on Pt utilization
- At lower RH, gravure coated CL losses more surface area
- Less ionomer-Pt contact

Measured by CO stripping voltammetry
H₂/N₂ Electrochemical Impedance Spectroscopy

\[ Z(\omega)_{\text{model}} = j\omega L_{\text{wire}} + R_\Omega + \frac{R_{CL}}{Q_{DL}(j\omega)^{\phi}} \coth \left( \sqrt{R_{CL} Q_{DL}(j\omega)^{\phi}} \right) \]

Setzler & Fuller, JES, 162 (6) FS19-FS30 (2015)
https://github.com/NREL/OSIF

100 %RH
0.2/0.2 sccm H₂/N₂
80 °C
V\text{dc} = 200 mV
V\text{ac} = 1 mV
1 – 10kHz

Δ\text{R}_{CL} = 33.1 \text{ m}\Omega \cdot \text{cm}^2
ΔE = i\Delta R_{CL}
ΔE \left( 0.8 \frac{A}{\text{cm}^2} \right) = 26.5 mV

Why does Gravure have higher resistance
• Higher tortuosity/fewer conduction pathways
• Lower surface ionomer content
Catalyst Layer Microscopy

- Gravure results in more clustered, denser catalyst layer
- Dense clusters lead to lower ionomer accessibility to catalyst – lower ECSA at low RH
- If ionomer is less dispersed it could lead to fewer proton conduction pathways
Catalyst Ink Rheology

- Catalyst inks are weakly agglomerated
- Shear thinning due to agglomerate break up
- Could the different shear rates of coating processes impact catalyst layer microstructure
How does coating method effect microstructure

**Slot Die**

- In Die Body – Plane Poiseuille
  - \( \dot{\gamma} = \frac{6Q}{W^2} \left(1 - \frac{y}{W}\right) \)
  - Avg \( \dot{\gamma} \approx 4 \text{ s}^{-1} \)
- In coating gap – Poiseuille/Couette
  - \( \dot{\gamma} = \frac{6Q}{LH^2} \left(1 - \frac{2y}{H}\right) + \frac{V}{H} \left(\frac{6y}{H} - 4\right) \)
  - Avg \( \dot{\gamma} \approx 50 \text{ s}^{-1} \)
- In tubing to die – Pipe Poiseuille
  - \( \dot{\gamma} = \frac{-4Qr}{\pi R^4} \)
  - Avg \( \dot{\gamma} \approx 110 \text{ s}^{-1} \)
  - Avg residence time in tube \( \approx 12 \text{ s} \)

**Gravure**

- Shear profile not analytically solvable
- Low shear in gravure cell

  [Image of shear profile]


- If we assume Couette flow between web and gravure lands and gap equals 2x liquid film thickness
  - Avg \( \dot{\gamma} \approx 300 \text{ s}^{-1} \)

*Suggests time at high shear is most important*
Conclusions

• Slot die leads to better dispersed catalyst particles which leads to more homogenous distribution of catalyst and ionomer
• This results in more ionomer in contact with catalyst, leading to better performance
• Indicates time at high shear, not just shear rate is important for high performance catalyst layers
Acknowledgements

NREL
• Min Wang
• Tim Van Cleve
• KC Neyerlin
• Mike Ulsh

Colorado School of Mines
• Samantha Medina
• Svitlana Pylypenko

• Nancy Garland
Thank You

www.nrel.gov

NREL/PR-5900-77815
Materials and Methods

- Catalyst: Pt on high surface area carbon (TKK TEC10E50E)
- Ionomer: Nafion, 1000 EW (Ion Power D2020)
- Membrane: Nafion, 1000 EW, 25 µm (Nafion NR211)
- Diffusion Media: Freudenberg H23C8
- Catalyst ink – 3.2 wt% PtHSC
- Ink Dispersion: High shear mixer (Ika Ultra Turrax)
- Gravure Coating
  - 1 m/min
  - Air floatation oven: 80 °C