Control of ionomer distribution and porosity in roll-to-roll coated fuel cell catalyst layers

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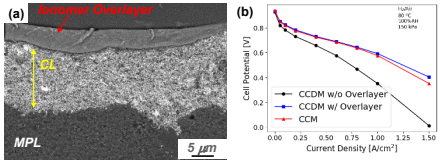
Objective

- Explore, understand and optimize material and process parameters for single-process (no extra ionomer over-layer) R2R manufacturing of GDEs with comparable performance to spray coated electrodes
- Can we create an R2R-coated electrode with an ionomer rich top surface?

Introduction

- Gas diffusion electrodes (GDEs) are of interest for roll-to-roll manufacturing due to easier handling of the substrate and compatibility issues between the ink and the hygroscopic membrane
- Spray-coated GDE require an additional layer of ionomer on top of the catalyst layer to match the performance of CCM electrodes



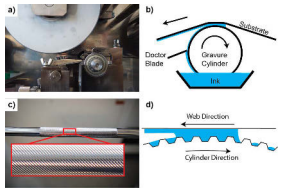
(a) TEM of image of a spray coated CCDM with ionomer overlayer. (b) Performance comparison between lab-scale spray-coated CCM baseline and GDEs with and without over-layer

Materials

- Catalyst – 50 wt% Pt on HSC (TKK TEC10E50E)
- Ionomer – Nafion 1000 EW (Nafion D2020)
- Membrane – 25 μm Nafion (NR211)
- GDL – SGL Sigracet 29BC
- Catalyst Inks
  - 3.2 wt% Pt/HSC
  - I:C – 0.9-1.6 w/w
  - Solvents: water and 1-propanol

Methods

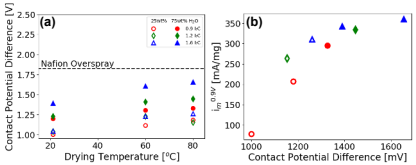
- Mayer rod
- R2R Coating
  - Microgravure
  - 1 m/min
  - 80 ˚C
  - 2 m of each ink
  - Loading: 0.1 ± 0.01 mg_{Pt}/cm^2
- Nano X-ray Computed Tomography
  - APS Beamline 32-ID
  - GDE's laser milled to 10 x10 μm²



a) Gravure coating station of NREL's R2R coating system. b) Illustration of gravure coating process. c) Gravure cylinder with zoom of tri-helical pattern. d) Illustration of fluid transfer process from gravure cylinder to substrate.

Results

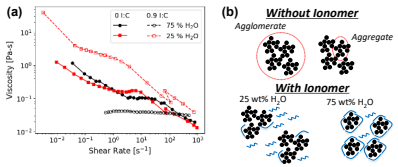
Characterization of Surface Ionomer Content



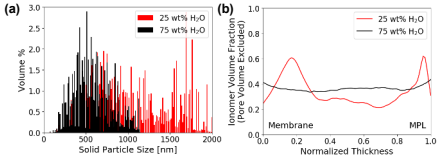
(a) Measured contact potential difference vs drying temperature for various ink formulations. (b) ORR mass activity vs cathode contact potential difference.

- Developed Kelvin probe method as non-destructive, rapid technique to quantify surface ionomer content
- Trends – increasing drying temperature and water content of ink increase surface concentration of ionomer
- Observed strong correlation between ionomer content and MEA performance

Influence of Solvent on Microstructure



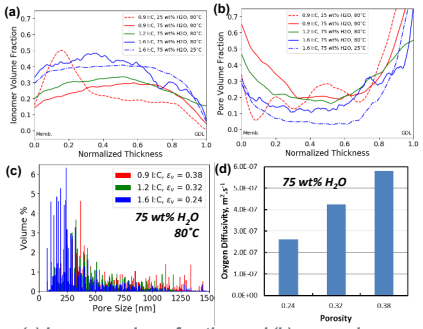
(a) Stead shear rheometry of Pt/HSC dispersions with (open) and without (solid) Nafion in water/1-propanol solvent blends. (b) Agglomeration of Pt/HSC without (top) and with ionomer (bottom)



XCT analysis of rod-coated 0.9 I:C electrodes. (a) Solid particle size distribution (b) Ionomer volume fraction as a function of layer thickness

- Water-rich solvent results in more dispersed ink due to different ionomer interactions
- More dispersed ink leads to smaller solid particles
- Stronger association of ionomer to catalyst leads to more uniform ionomer distribution

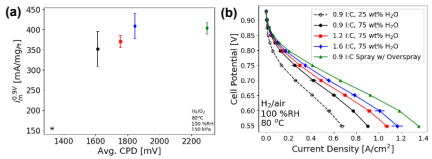
XCT Analysis of Ionomer Distribution and Porosity



(a) Ionomer volume fraction and (b) pore volume fraction profiles for different ink formulations and catalyst layer drying conditions. (c) Pore size distribution for catalyst layers cast from water-rich ink and (d) resulting oxygen diffusivity.

- XCT verifies Kelvin probe measurements
- Higher drying temperature increases porosity
- Increasing I:C decreases pore size and oxygen diffusivity

Roll-to-Roll Coated GDEs



(a) ORR mass activity vs cathode contact potential difference. (b) Air polarization curves of R2R-coated GDEs compared to spray-coated GDEs

- Demonstrated R2R electrodes without ionomer overlayer produce equivalent mass activity to spray-coated electrodes with ionomer overlayer
- R2R GDEs have lower high-current density performance about 14% than spray-coated GDEs

Conclusions

- Ink formulation and drying parameters can be tuned to control the catalyst layer ionomer distribution and porosity
- Future work focused on greater control of ionomer distribution using different solvents

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