High-Performing and Durable Electrodes for PEMFCs

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Introduction

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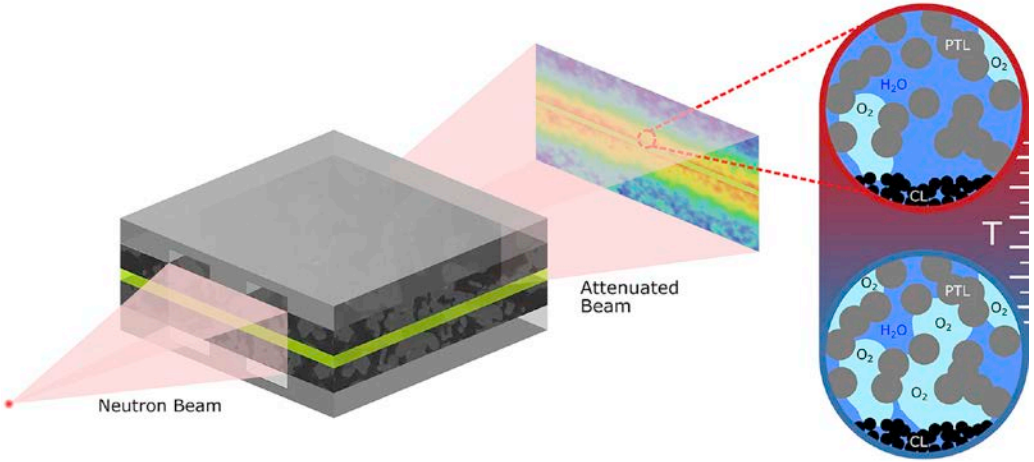
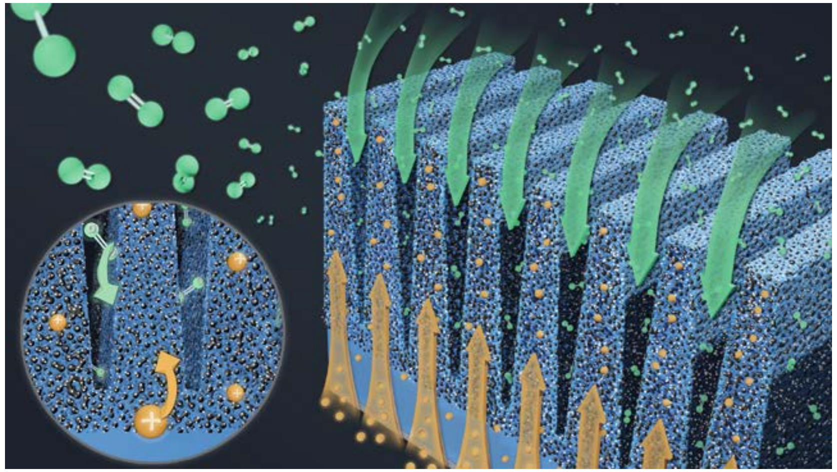
Flow and Mass Transport for PEM Electrolyzers



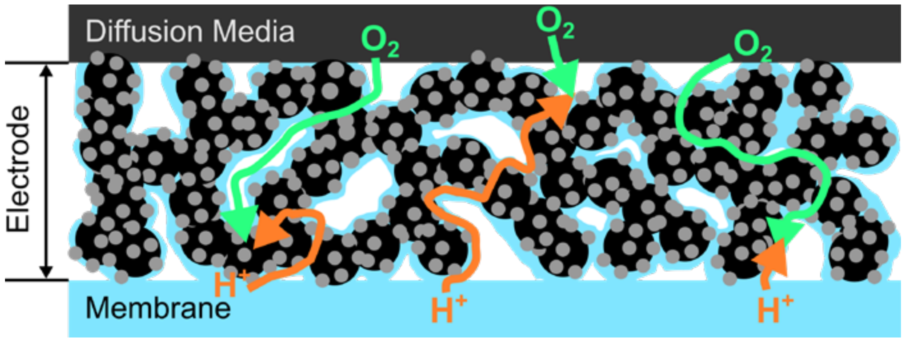
High-Performing and Durable Electrodes for PEMFCs



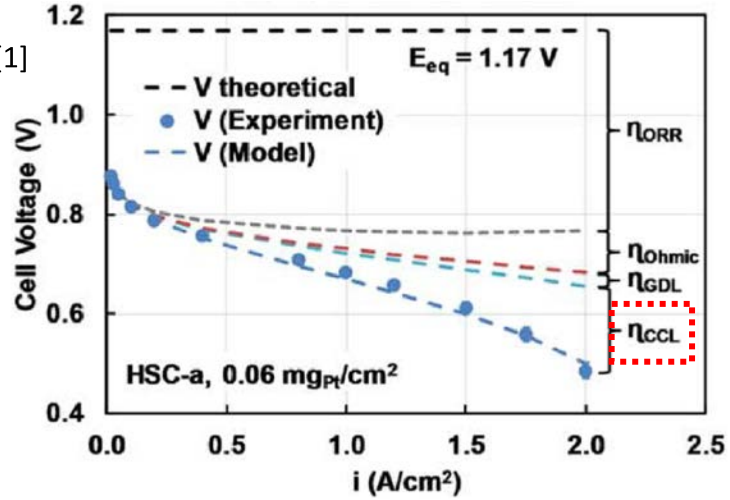
#1: Next Generation Electrode Structures for PEMFC

Motivation: challenges for conventional electrodes

- Random mixture of Pt/C, ionomer, and pore
- Tortuous and inefficient H^+/O_2 transport pathways

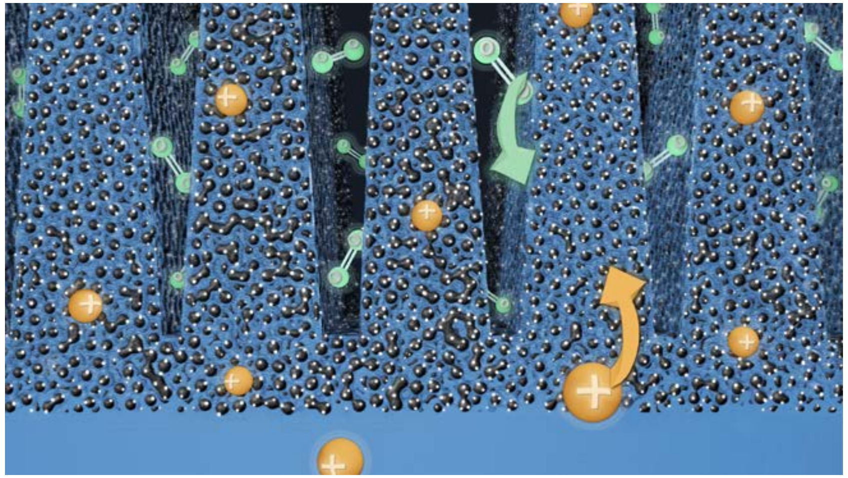


[1]



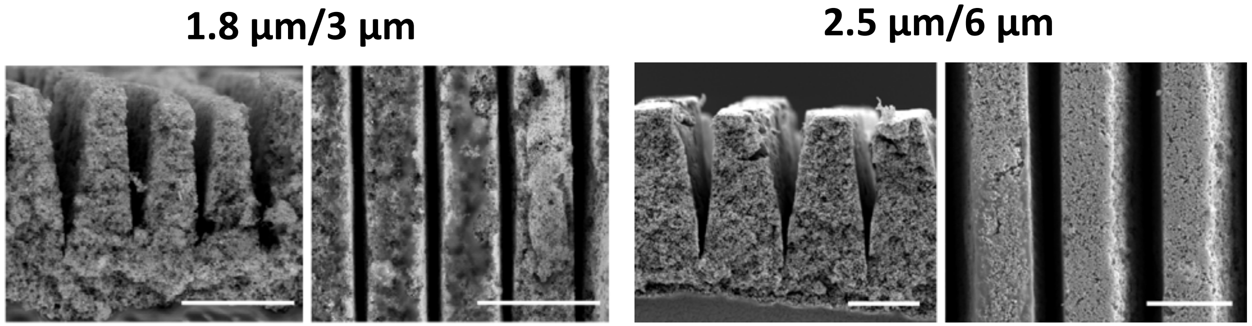
Approach

- Partition H^+/O_2 transport pathways via **groovy electrodes**



Two main features

1) High I/C electrode ridges for H^+
2) Grooves for O_2

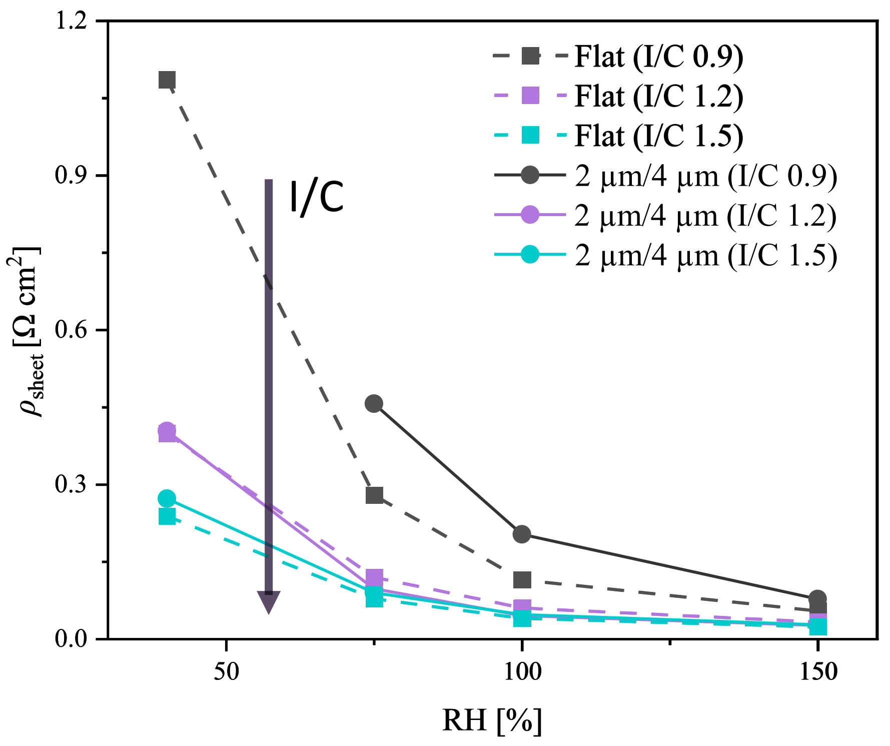


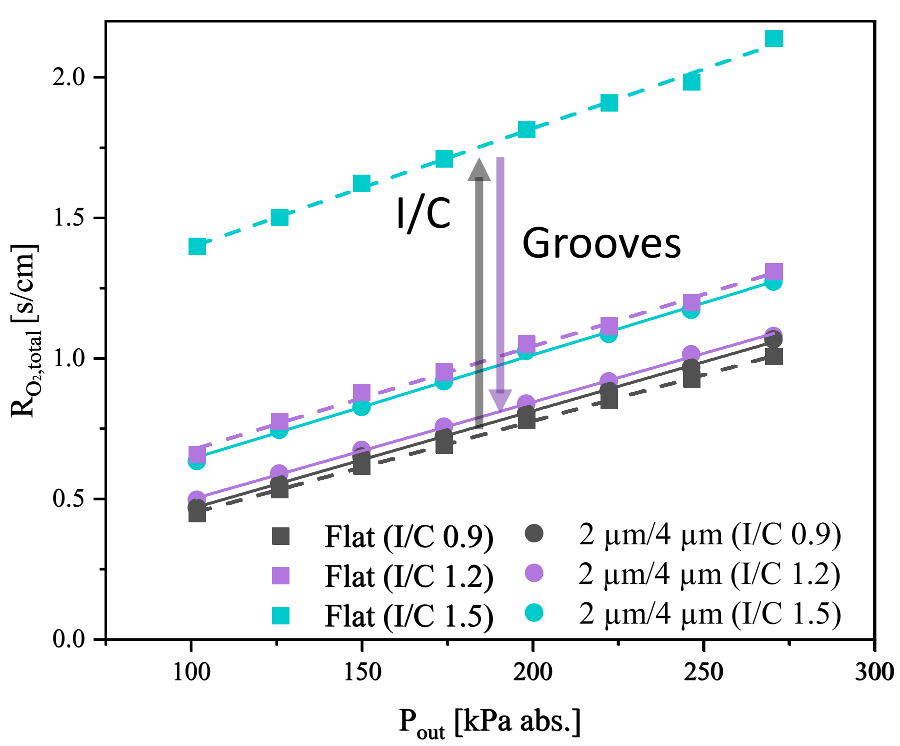
Scale bar: 5 µm

Groove Width/Groove Spacing

[1] Ramaswamy et al., Journal of Electrochemical Society, 167, 064515 (2020)

Groovy Electrodes Enable Facile H^+/O_2 Transport

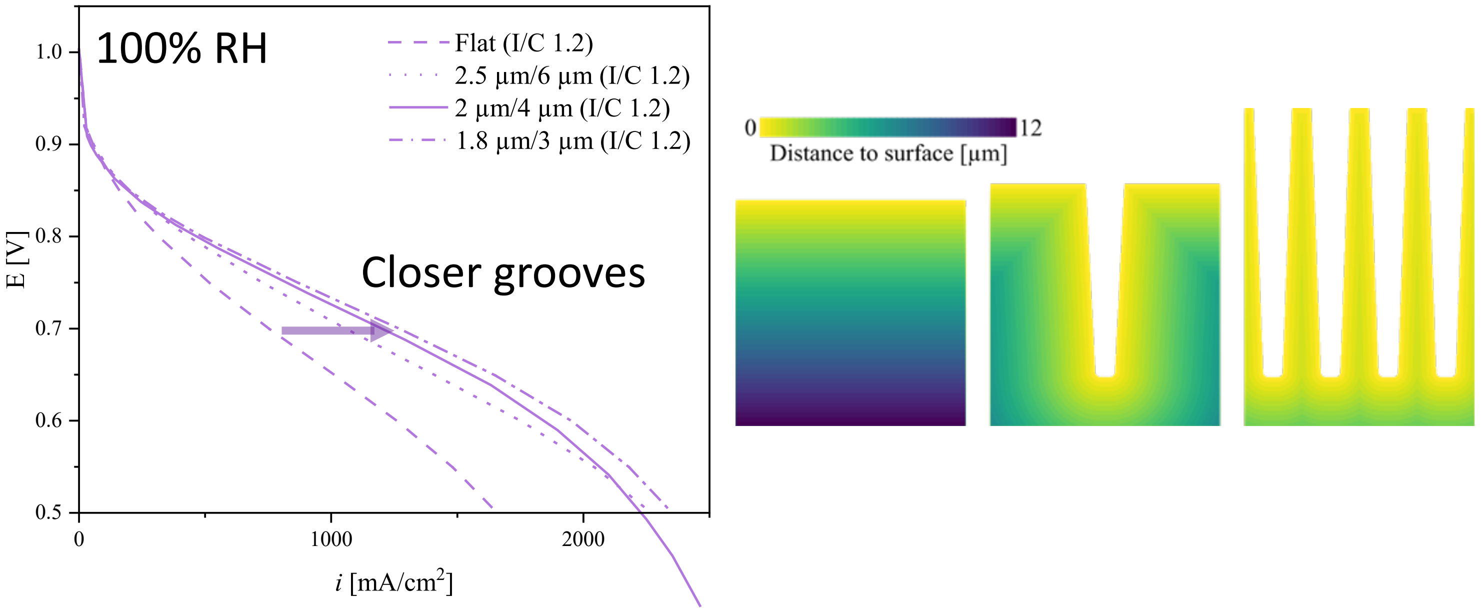




- Increase in I/C ratio reduced sheet resistance.
- Similar trend was observed after the addition of grooves
- 60% decrease (groovy I/C 1.2 vs. flat I/C 0.9)

- Increase in I/C ratio increased oxygen transport resistance.
- R_{O2} of groovy (I/C 1.2) became comparable to that of flat (I/C 0.9).

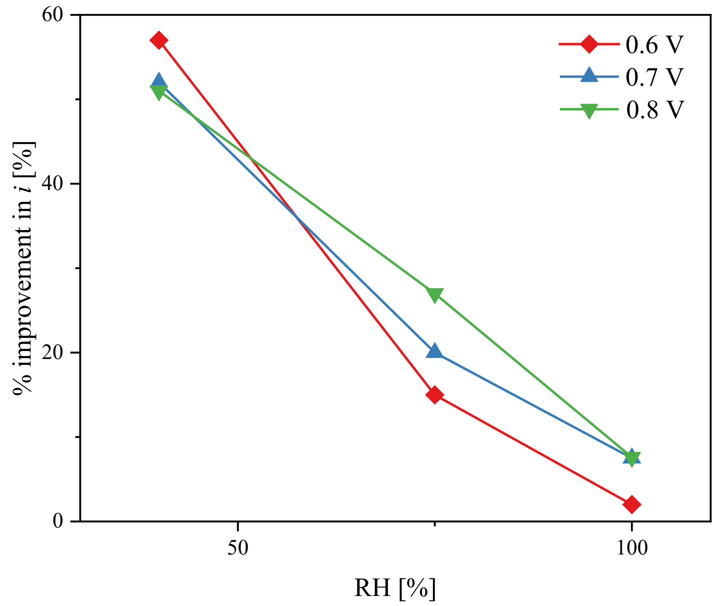
Closer Grooves Enhance Performance



- Closer grooves led to **shorter O_2 diffusion path**.
  - Reduction in groove spacing leads to reduced distance from electrode interior to surface
  - 8-fold from flat to 1.8 µm/3 µm

Cell: 0.3 mg_{Pt}/cm^2, I/C 0.9, TEC10E40E, N211, SGL 22BB
Testing: 5 cm^2 differential, 1000/3000 sccm H_2/Air, 150 kPa, 80°C

Flat (I/C 0.9) vs. 1.8 µm/3 µm (I/C 1.2)

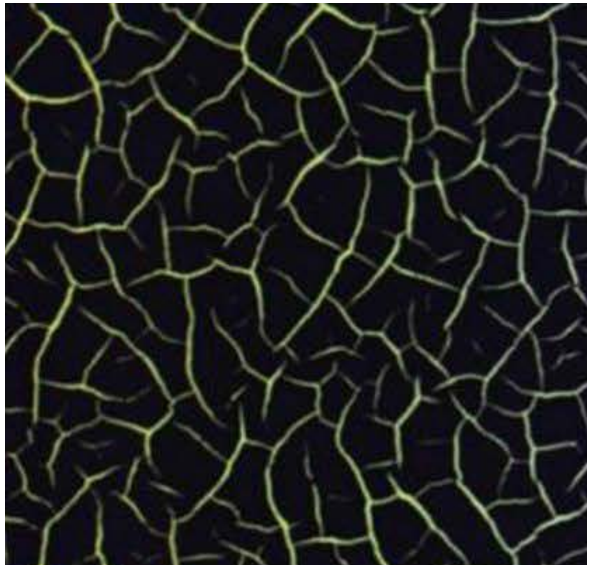


- Improved performance across a wide range of operating conditions
- Groovy electrodes are particularly advantageous under **dry** conditions.
  - H^+ transport-limited under dry conditions, and higher I/C enhances H^+ transport.

#2: Electrode Crack Density Effects on Durability

Motivation: challenges of crack investigation

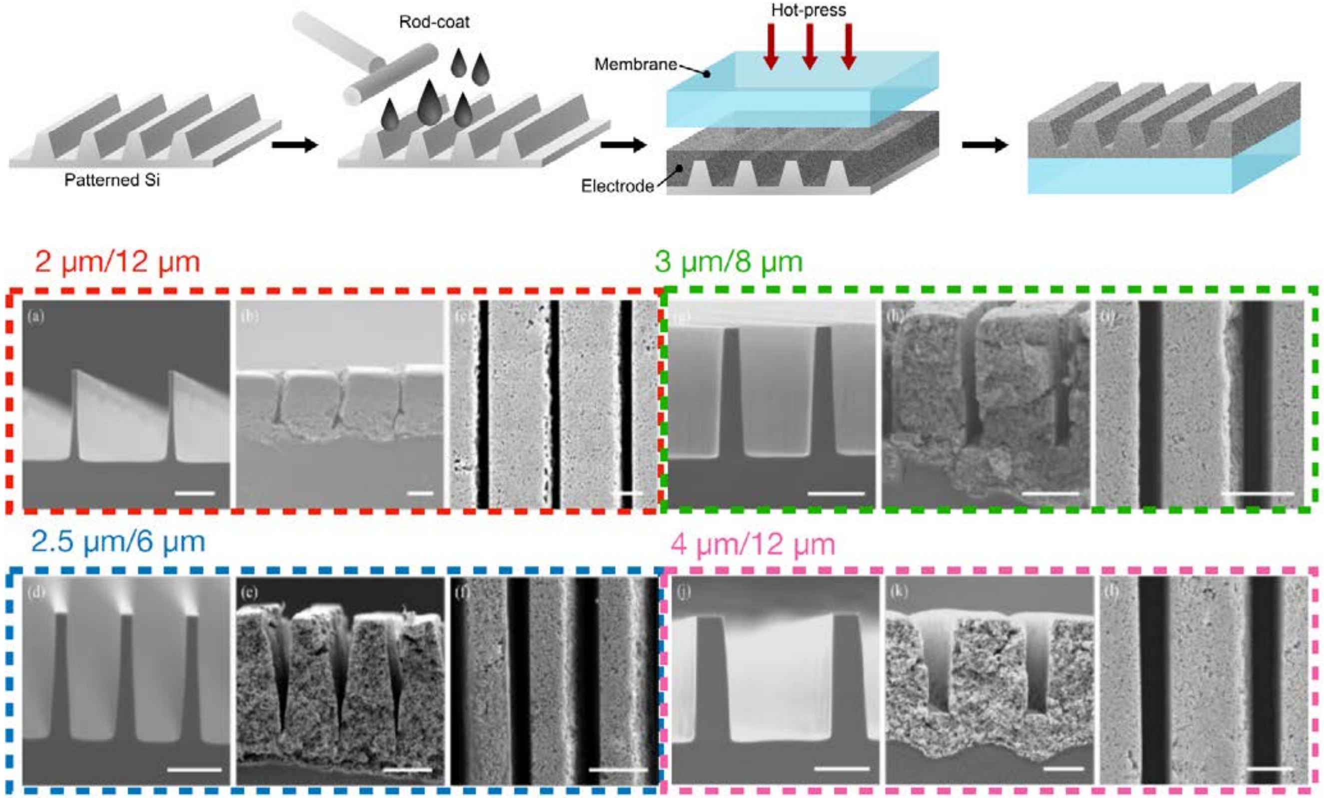
- Difficulty in controlling crack width, depth, density, and orientation
- Unclear effects of cracks on electrode durability



[1]

Approach

- Using lithography-based approach to engineer cracks with prescribed morphology

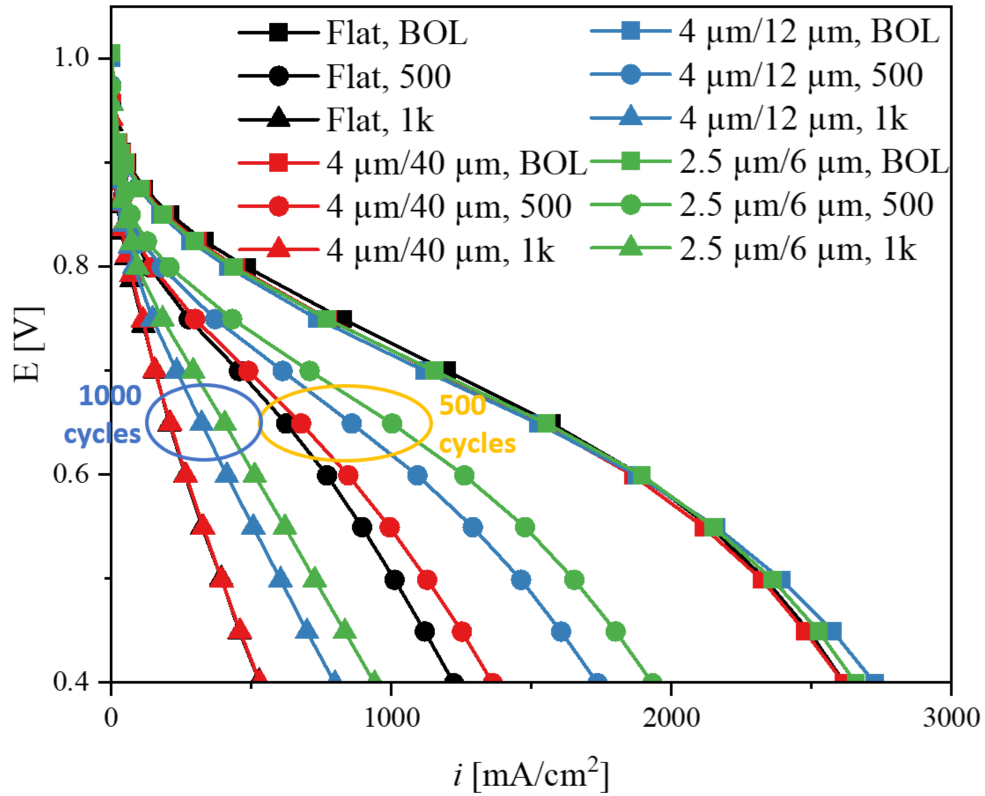


Scale bar: 5 µm

Crack Width/Crack Spacing

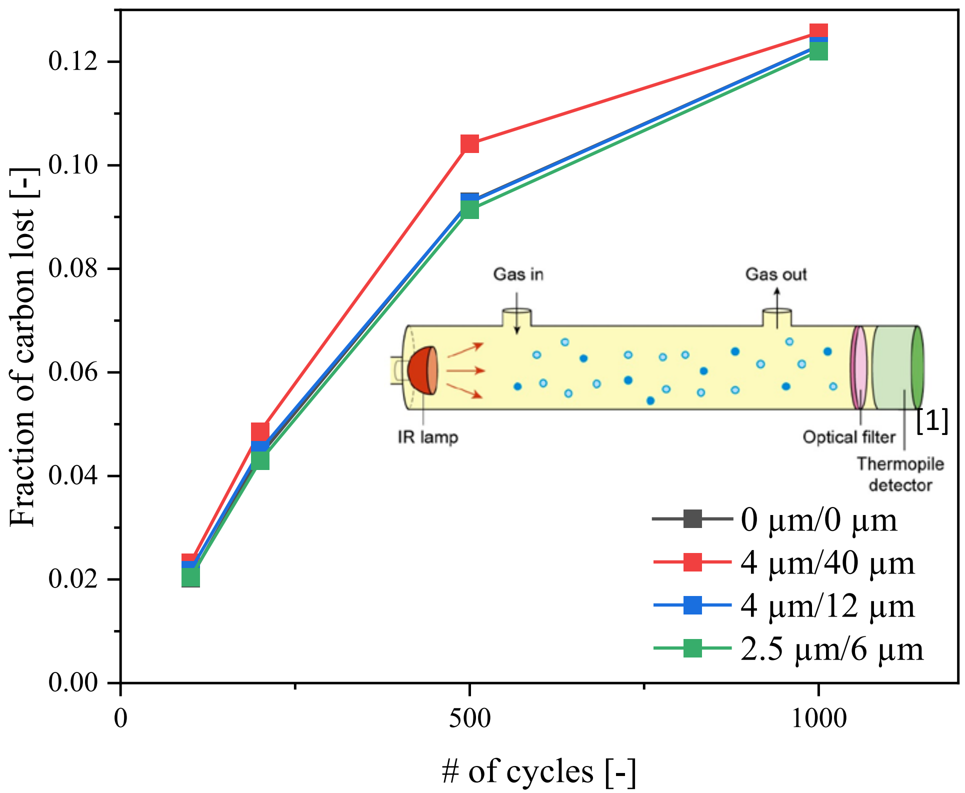
[1] Kumano et al., Journal of Power Sources, 419, 219 (2019)

Denser Cracks Improve Performance Post-AST



- After support AST, denser cracks **enhance** the performance of PEMFCs.

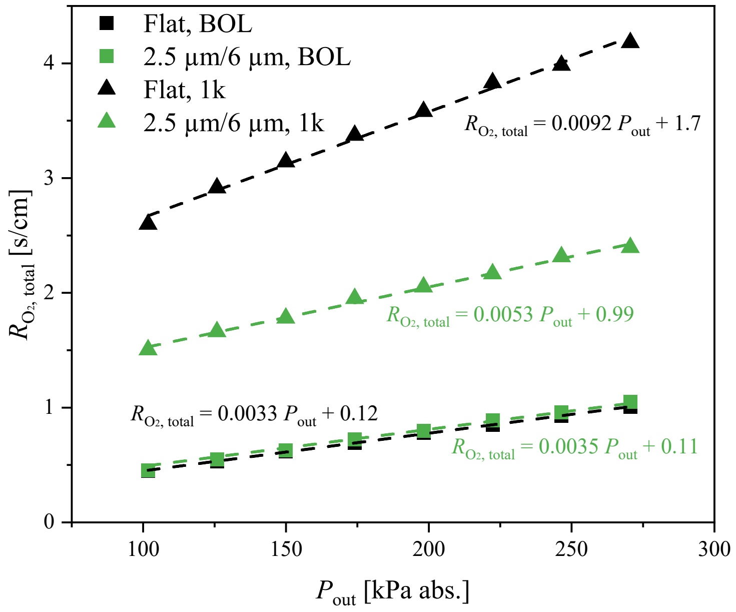
Cell: 0.3 mg_{Pt}/cm^2, I/C 0.9, TEC10E40E, N211, SGL 22BB, I/C 0.9
Testing: 5 cm^2 differential, 1000/3000 sccm H_2/Air, 150 kPa, 80°C, 100% RH

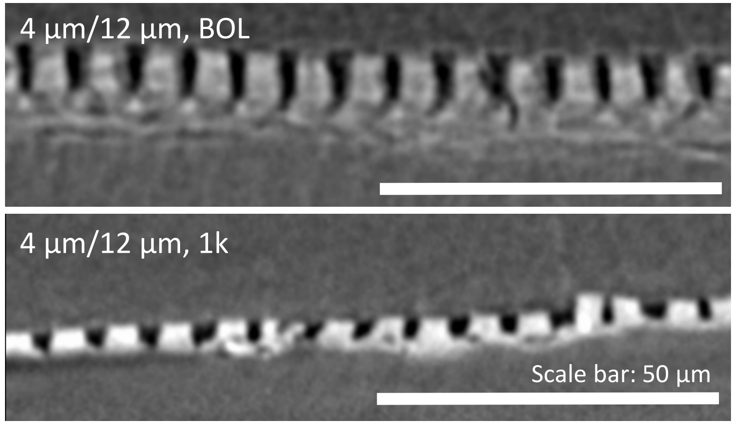


- **Negligible difference** in carbon loss (measured via NDIR), MA, ECSA, and HFR.

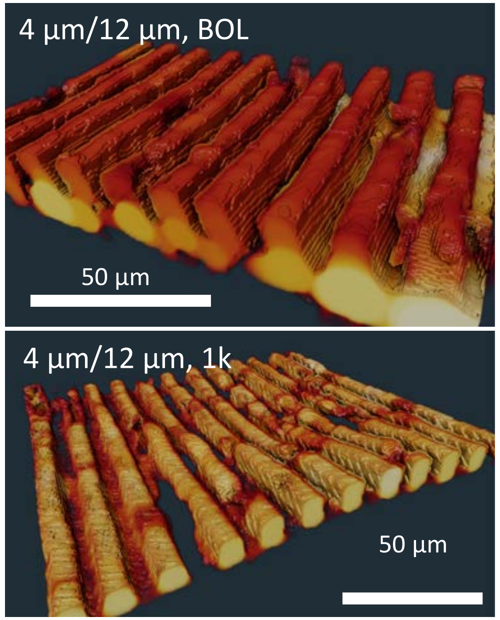
[1] Mendes et al., Sensors, 15, 11239 (2015)

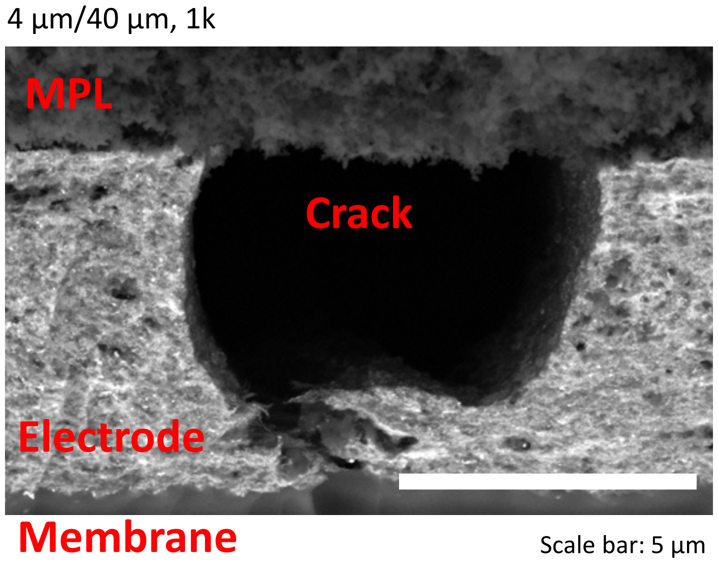
Electrode Cracks Reduce O_2 Transport Resistance





- After support AST, R_{O2} is significantly **lower with cracks**.
  - ➢ Electrode collapses, leading to R_{O2}-limited structure
  - ➢ Cracks provide shorter O_2 path
- Cracks are getting wider with carbon corrosion.
  - ➢ **Preferential corrosion near cracks?**





- **No preferential corrosion** was observed.

- Two hypotheses:
  1. Cracks mainly act as O_2 pathway rather than H_2O
  2. Support AST conditions do not flood the cracks

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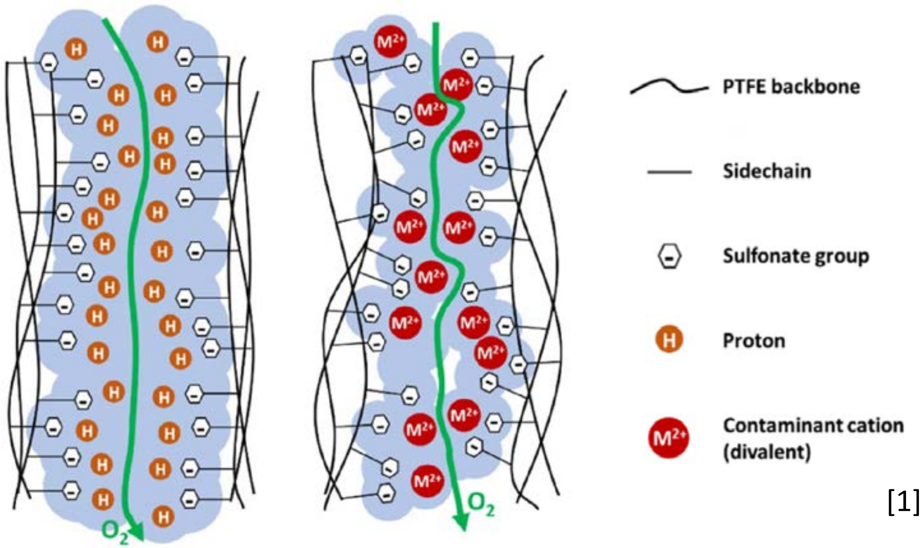
#3: Cobalt Contamination in PEMFC Electrodes

Motivation: challenges of understanding Co^{2+} effects

- Deconvoluting physical loss of Co and effect of Co^{2+} contamination in ionomer/membrane



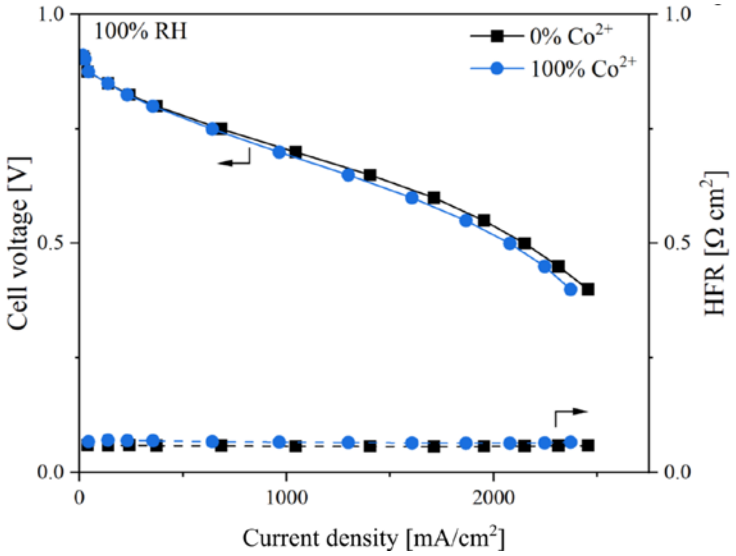
[1]

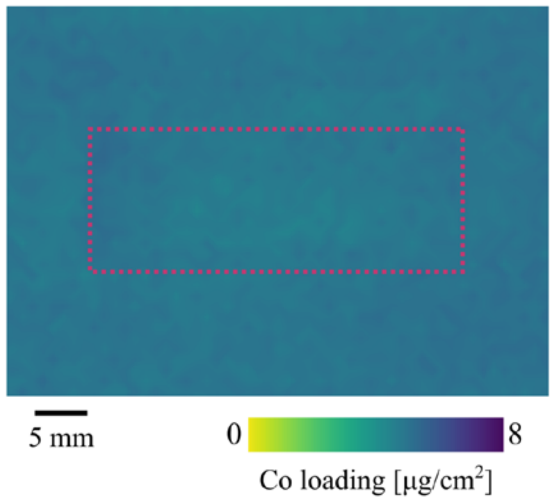
M2FCT Year 1 Milestone (Q3):

Acceptable transition metal loss from alloy catalysts (% of sulfonic acid sites in ionomer layer) defined with respect to electrode layer losses. (LBNL, LANL, ORNL, ANL, NREL)

Approach

- Electrode decal-based MEAs tested under various % Co^{2+}





- Co^{2+} effects were not observed during initial investigations.
- **Inactive membrane area** acts as a Co^{2+} sink, which suppresses Co^{2+} effects on performance.

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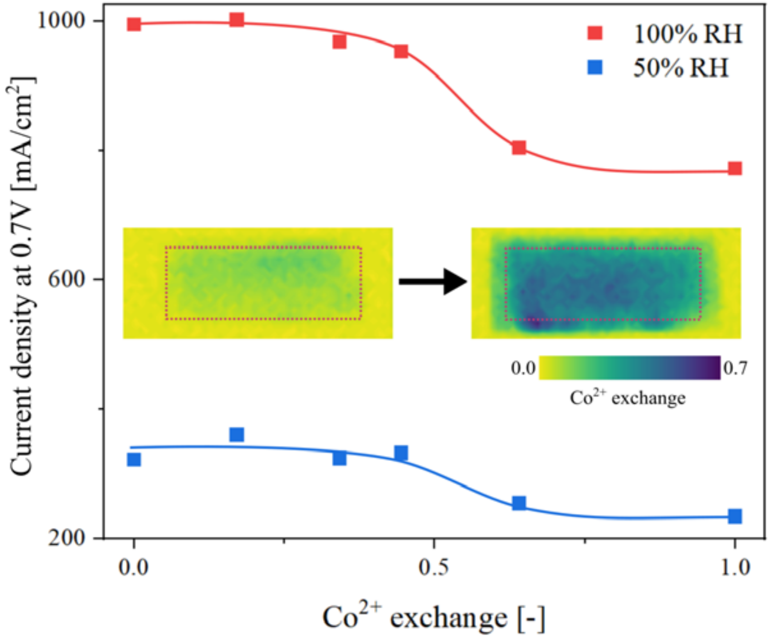
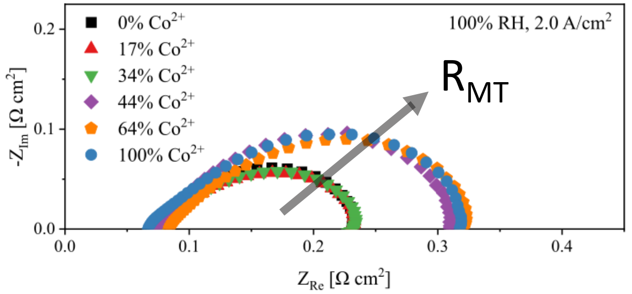
[1] Braaten et al., Journal of Electrochemical Society, 166, F1337 (2019)

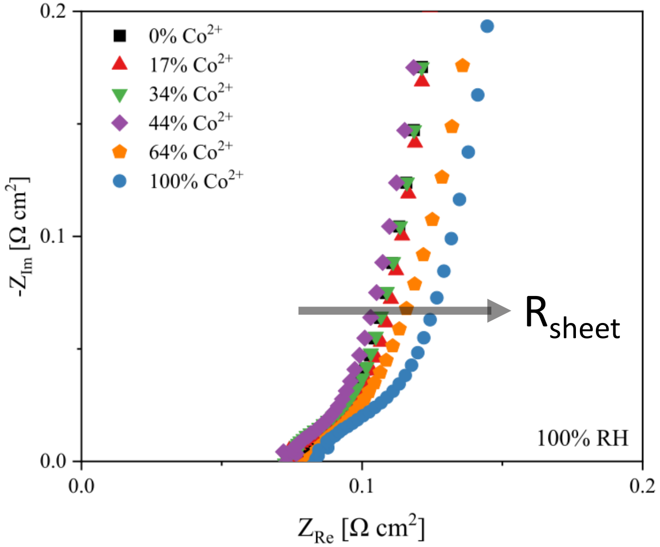
Cobalt Contamination in PEMFC Electrodes



~44% Co^{2+} exchange acceptable

- R_{MT} **increased** with increasing Co^{2+} doping.
  - Water uptake in the ionomer decreases with increasing Co^{2+} doping.
- R_{sheet} **increased** with increasing Co^{2+} doping.
  - Co^{2+} ion-exchanging with sulfonic acid sites lead to poor proton transport.
- Membrane resistance, and kinetic resistance remained relatively unchanged.





Cell: 0.25 mg_{Pt}/cm^2, I/C 0.9, TEC10E40E, N211, SGL 22BB, I/C 0.9
Testing: 5 cm^2 differential, 1000/3000 sccm H_2/Air, 150 kPa, 80°C

Summary and Future Work

#1: Groovy Electrode Enhances Performance.

- Enhanced H^+ and effective O_2 transport led to improved performance, particularly under dry conditions.
- **Future work** will couple experimental results with computational tools to further optimize structure for enhanced performance.

#2: Electrode cracks lead to improved performance after support AST.

- After electrode collapses, cracks act as a short-cut for O_2 to diffuse to reaction sites.
- **Future work** will examine the effect of electrode cracks on membrane durability during RH cycling.

#3: ~44% Co^{2+} exchange can be tolerated in electrode ionomers.

- Co^{2+} exchange results in higher R_{MT} and R_{sheet}.
- **Future work** will explore operating strategies/electrode designs to suppress Co^{2+} effects.

Aspirations: develop my own research group centered around hydrogen and fuel cell technology, to (1) advance the science and technology and (2) train next generation scientists and engineers.

Acknowledgements

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- Membrane measurements by Adlai Katzenberg, Ahmet Kusoglu (**LBNL**)
- Impedance modelling by Xiaohua Wang, Jui-Kun Peng, Rajesh K. Ahluwalia (**ANL**)

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