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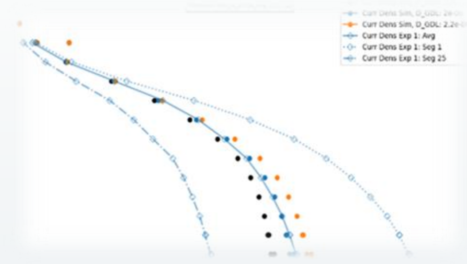
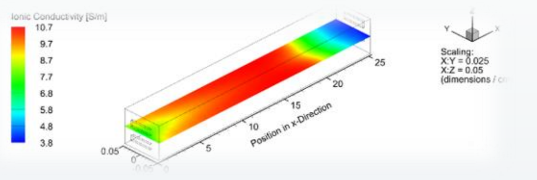




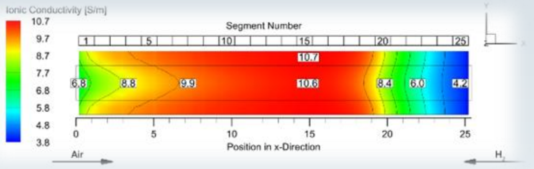
Spatially resolved simulation on PEM fuel cells meets experiments – which model complexity is enough under which conditions?

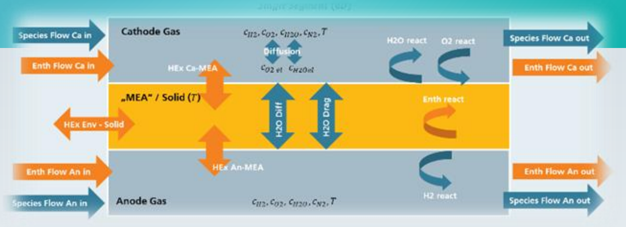
T. Kurz, J. Schall, N. Raue, D. Gerteisen
Department Fuel Cell Systems
Fraunhofer Institute for Solar Energy Systems

FDFC, 25 Sep 2023, Ulm, Germany

www.h2-ise.com

www.ise.fraunhofer.com



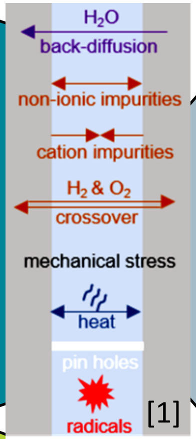


PEMFC Modeling: Many coupled physical processes

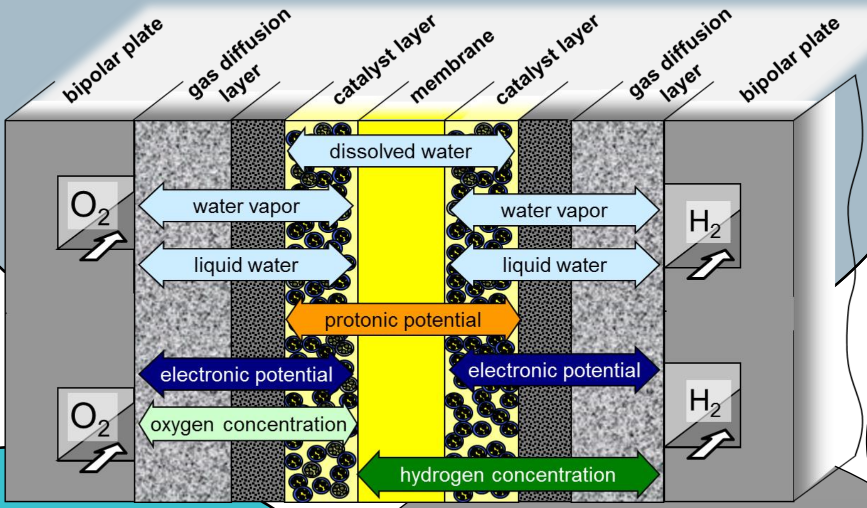
Which processes are important for sufficient results?

Membrane related

Absorption, Desorption, Diffusivity, Osmotic Drag, Conductivity, Chemical and Mechanical Stability…

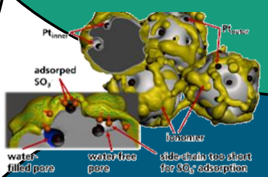


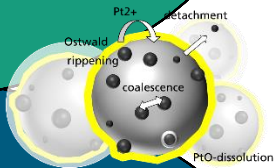
Many Domains, Sources and Sinks (Heat, Charge, Species)



Dynamics and Ageing

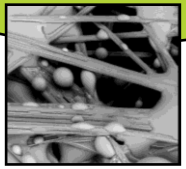
Poisoning, Degradation, Side chain restructuring, Catalyst (restructuring) and Oxidation, Carbon Corrosion…





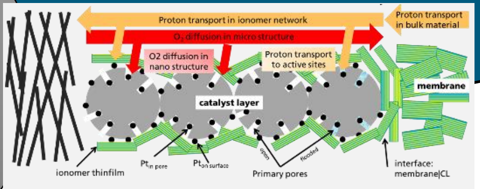
Water related

Humidity, Activity, 2-Phase Flow, Condensation, Evaporation, Capillary Effects…



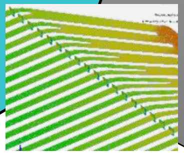
Electrodes

Structure, Composition, 3-P-Boundaries, Species Transport, Reaction, Kinetics, Electric and Ionic Conduction, Catalytics, Poisoning, Degradation…



Transport of

Gases, Liquid, Dissolved Species, Heat, Charge…



Transport in

…Channels, Pores / Fibres, Solid, Ionomer…

…with Convection, Diffusion, Conduction…

[1] E. Dickinson, G. Smith, Membranes 2020, 10, 310; doi:10.3390/membranes10110310

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Model geometry: Relevant dimensions

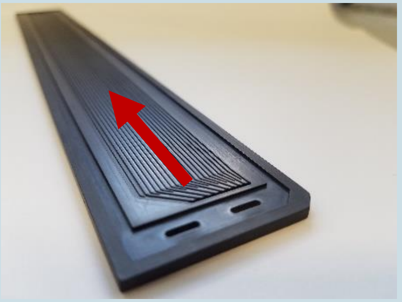
Three main directions

Relevant processes and model dimension depend on

- Model aim / focus
- Required accuracy
- Desired calculation time

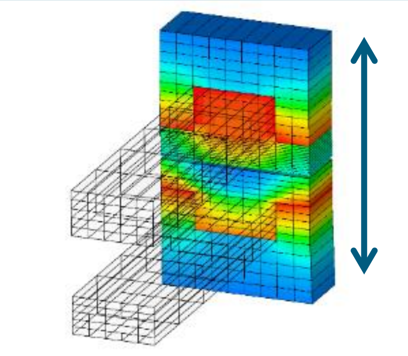
Along The Channel (ATC)

- O2 / H2 concentration gradient due to depletion
- Humidity increase along air flow
- Humidity and conductivity gradient
- Temperature gradient
- Pressure loss



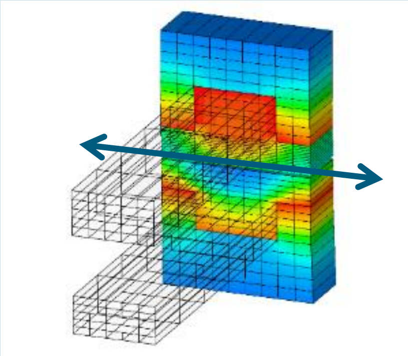
Through Plane (TP)

- 7+ Domains: (FFP) / Ch / GDL / El / M / El / GDL / Ch / (FFP)
- Strong gradients of concentration, humidity…
- ATC 1D does not account for TP gradients, process variables at the interfaces are avg values



Channel / Rib (CR)

- Different tortuosity for gas and water transport leads to differences in humidity and gas concentrations
- drying under channel or flooding under rib possible
- Area effective values for e.g. diffusion coefficients

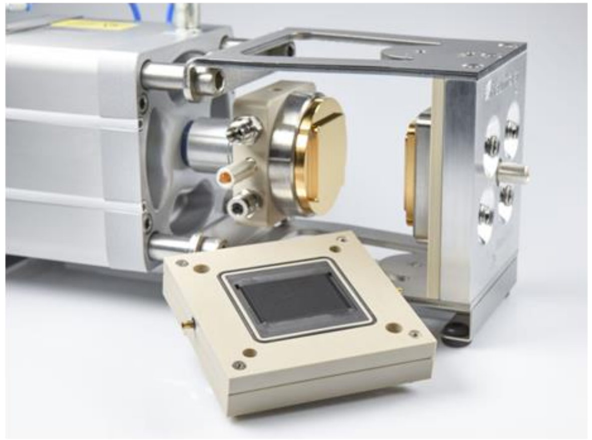


Along-the-Channel testing and modeling

Current and impedance characteristics along the gas channel

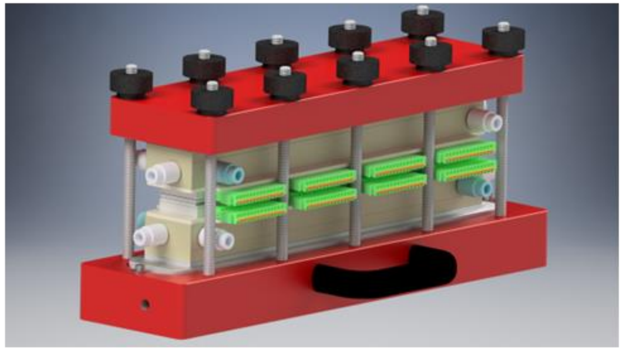
Differential cell *(D)*

- Local spot inside the full size cell
- Small active area (here: 12 cm²)
- "No" gradients
- Material testing



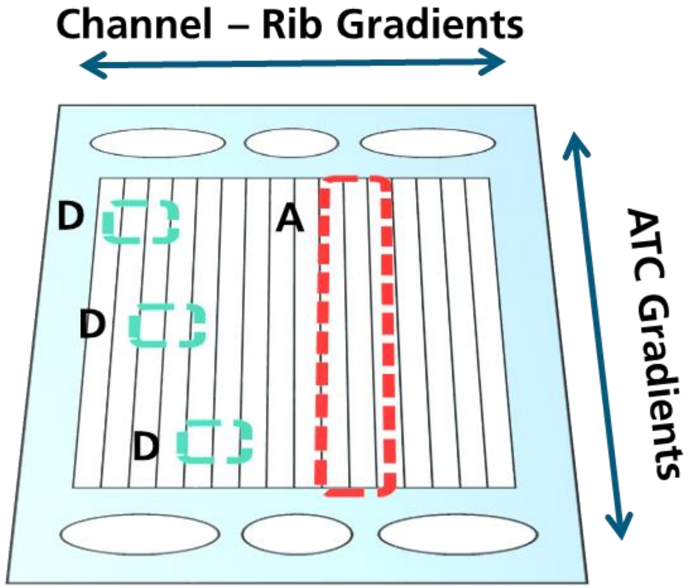
ATC cell *(A)*

- Section of full-size cell in one channel dimension
- Full channel length (real Δp)
- 1D gradients



Full size cell

- Automotive size (250+ cm²)
- 2D gradients (c, RH, T, I, V,…)



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Along-the-Channel Test Cell @ Fraunhofer ISE

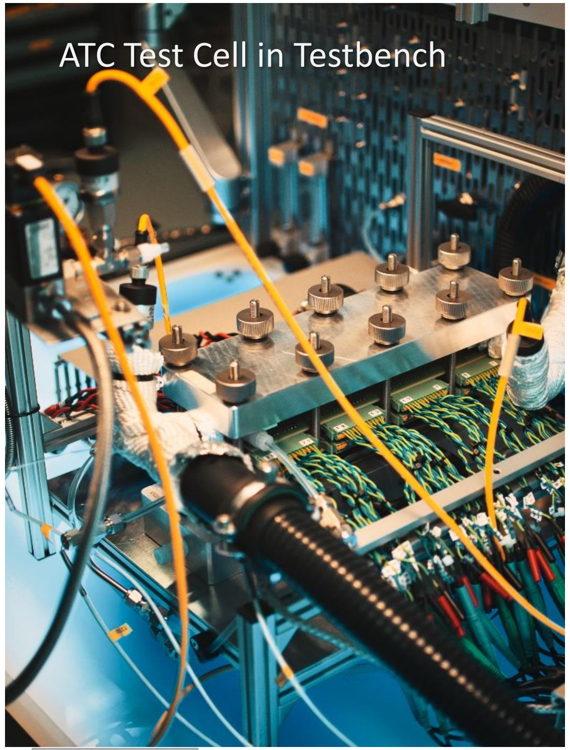
Analysing Current and Impedance Characteristics Along-the-Channel

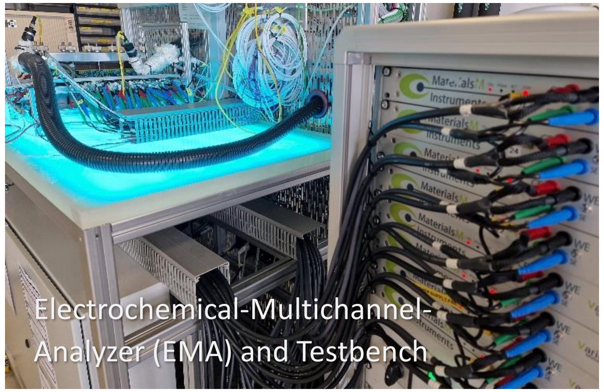
Cell characteristics

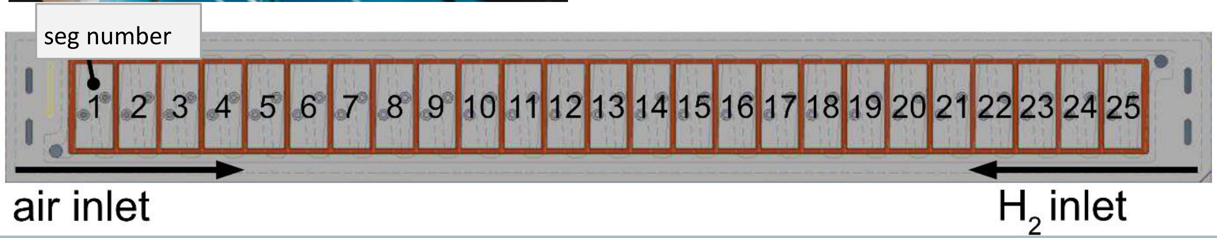
- 250 x 20 mm active area
- Parallel gas channels
- Plates separated in 25 electr. insulated segments
- Electric heating, liquid cooling on both sides
- Co-flow and counter flow possible

Test Setup

- Controlled humidification, temperature, pressure and mass flows
- Each Segment connected to a separate power potentiostat for current distribution, CV and EIS measurements





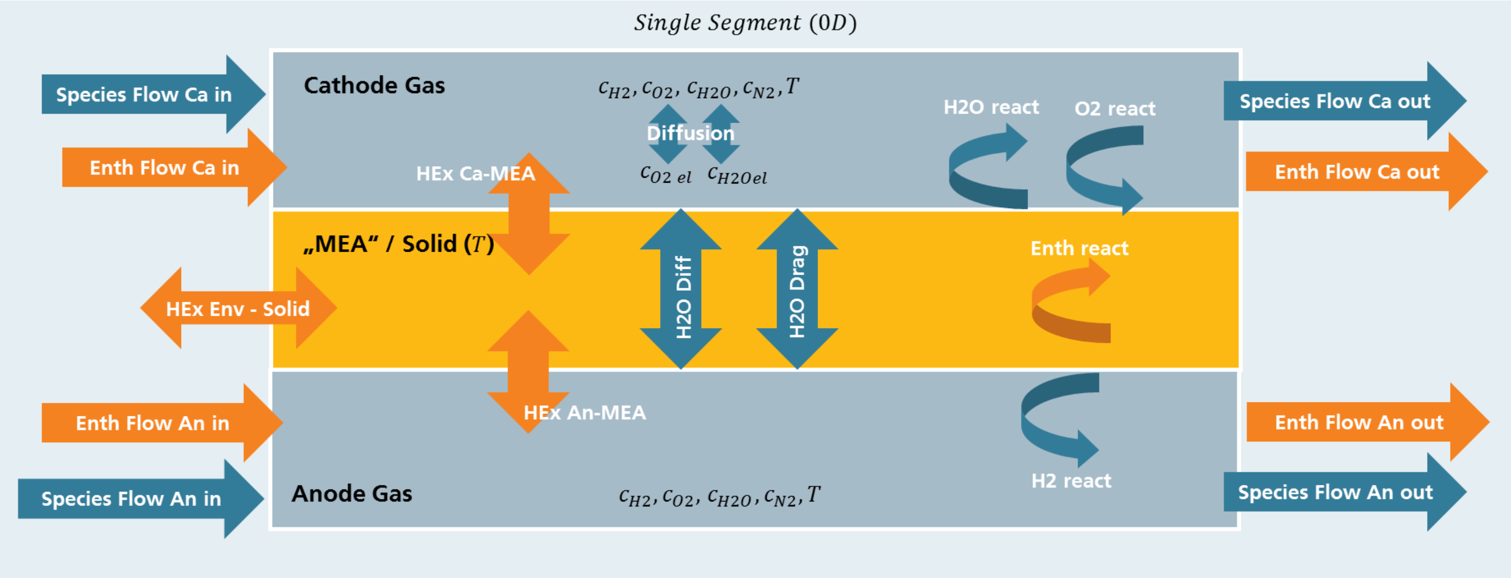


ATC Model – Deeper understanding of ATC measurements

Single Segment

Aim: Supporting ATC measurements

- Fast analysis
  →"simple" model required
- Complex enough to help analysing ATC results
- Dynamic and non-isothermal
- Flexibility and modularity
  - Number of segments, adding / removing processes (balance equations), co flow / counter flow, gal / pot mode), exchange CCM model

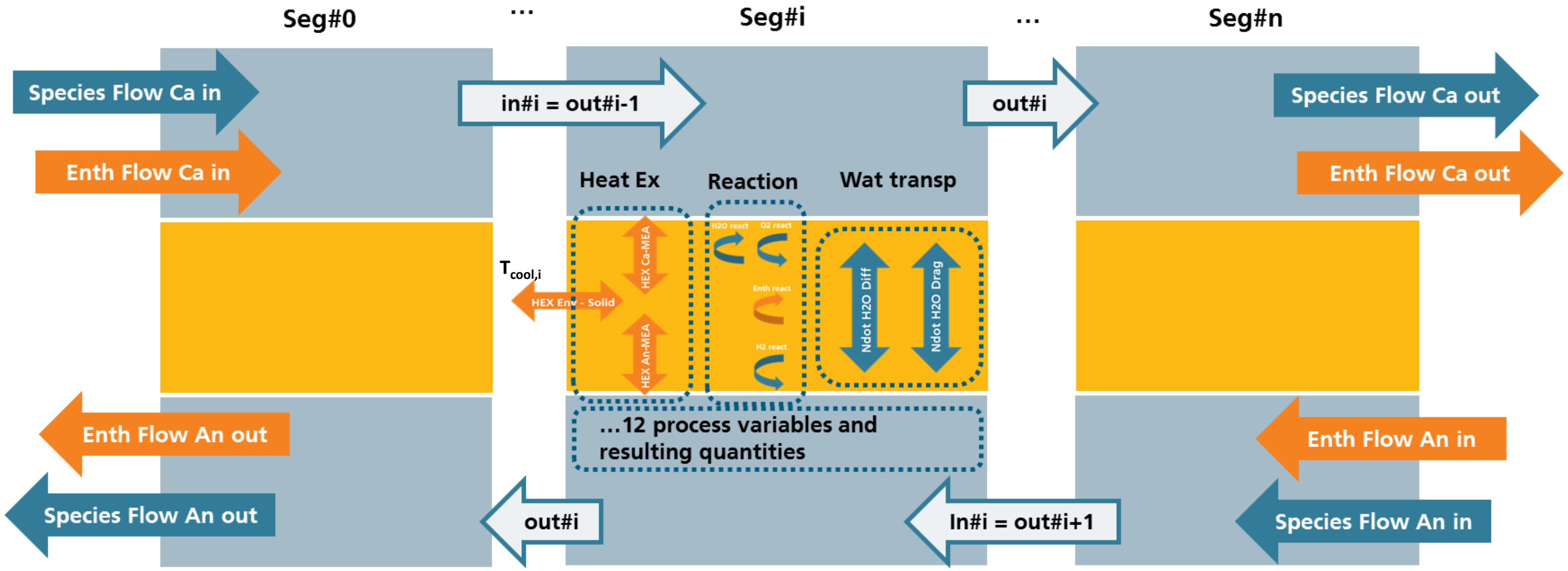


- **3 Domains, 12 Process Variables,** 4 Species, Mass- and Energy Balance, Tafel Approach (Cathode)
- BCs: Anode Inlet, Cathode Inlet, Total Pressure and Ambient Temperature at each Segment

ATC Model: 0D → 1D

Specifications and Limitations

- Coupling N segments along the gas channel
- Process variables are dependent on previous / following segments
- Coupled solution (300 balance equations @25 Segments)
- Implemented in python (numpy solve_ivp)
- All boundary conditions from ATC cell



- **Many Limitations:** Serial coupling of 0D-Segments, no channel / rib distinguishing, no liquid water, no through plane resolution, simple Tafel approach (only cathode), electrode is interface, diffusion only through cathode GDL
- **Fast Calculations:** Time to reach stationary solution (on „default“ i7 PC)
  - 0.5…1 s for 5 Segments
  - 5…15 s for 25 Segments

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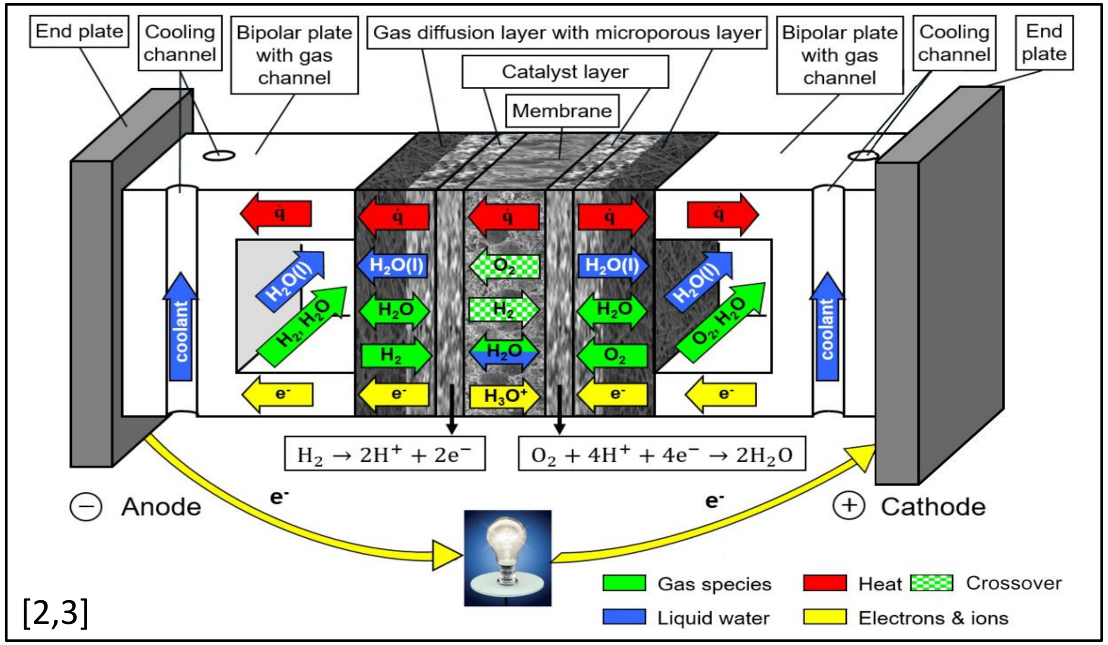
3D CFD Simulation (AVL Fire M)

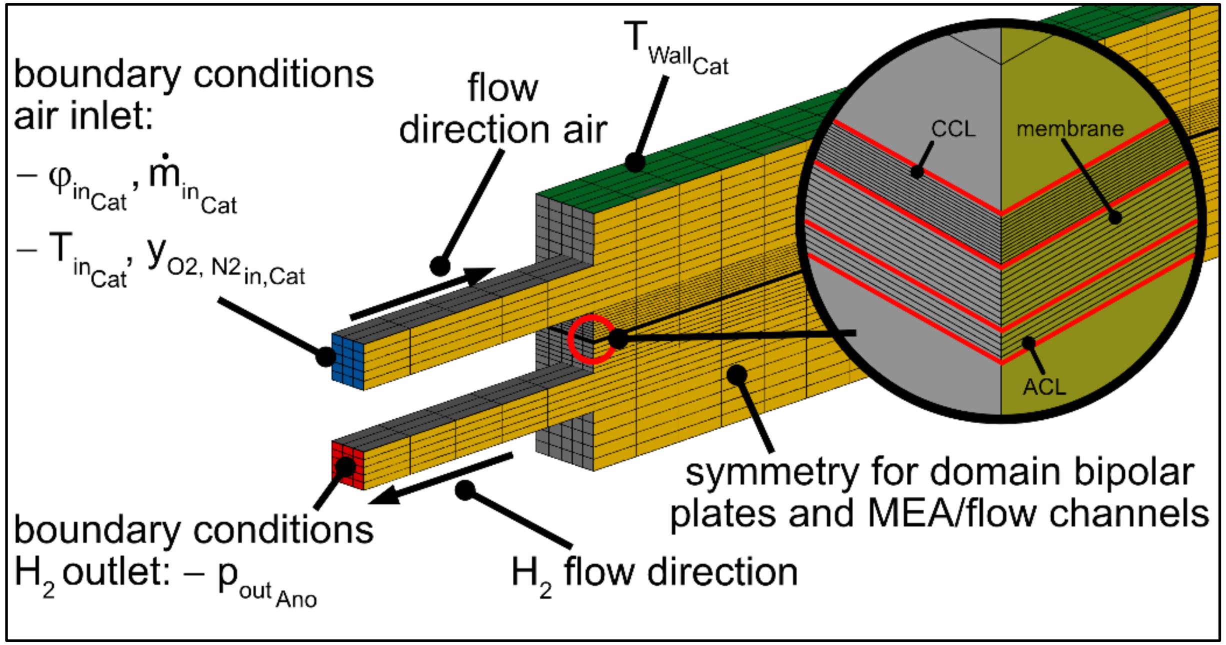
Model characteristics

Geometry and Boundary Conditions

- Simulation of a half single-channel to reduce calculation time: ~210k cells: ~5 hours for stationary solution
- Domains: Bipolar plates, Membrane, Porous Electrodes, GDLs
- Cooling channels replaced by boundary conditions (T_{Wall})
- Operating conditions from experiment



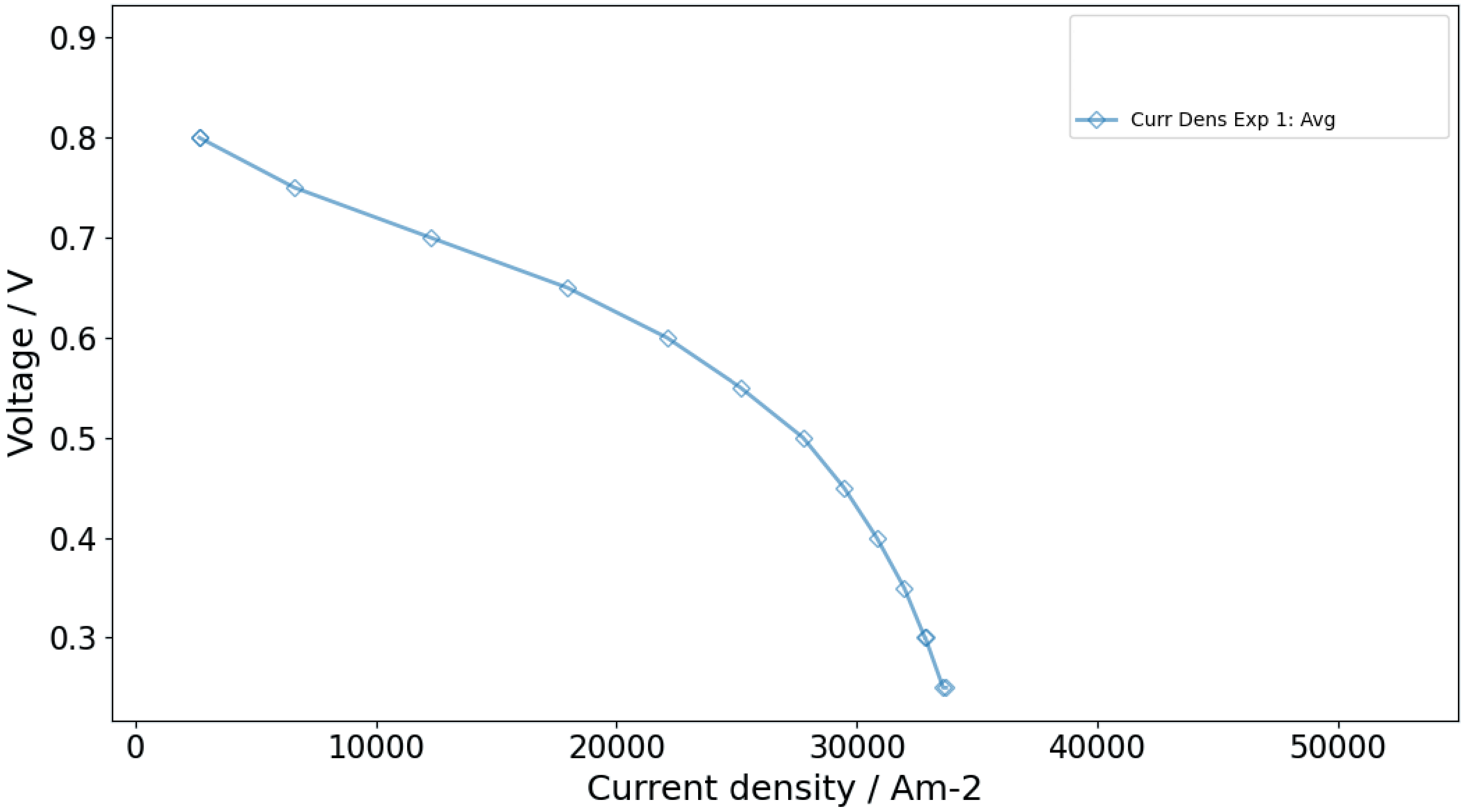
[2,3]



[2] AVL FIRE M Manual R2022.1
[3] Fink et al., Fuel Cells 20, 2020, No. 4, 431–452 2020.

Experimental ATC Results
Polarisation Curve Validation

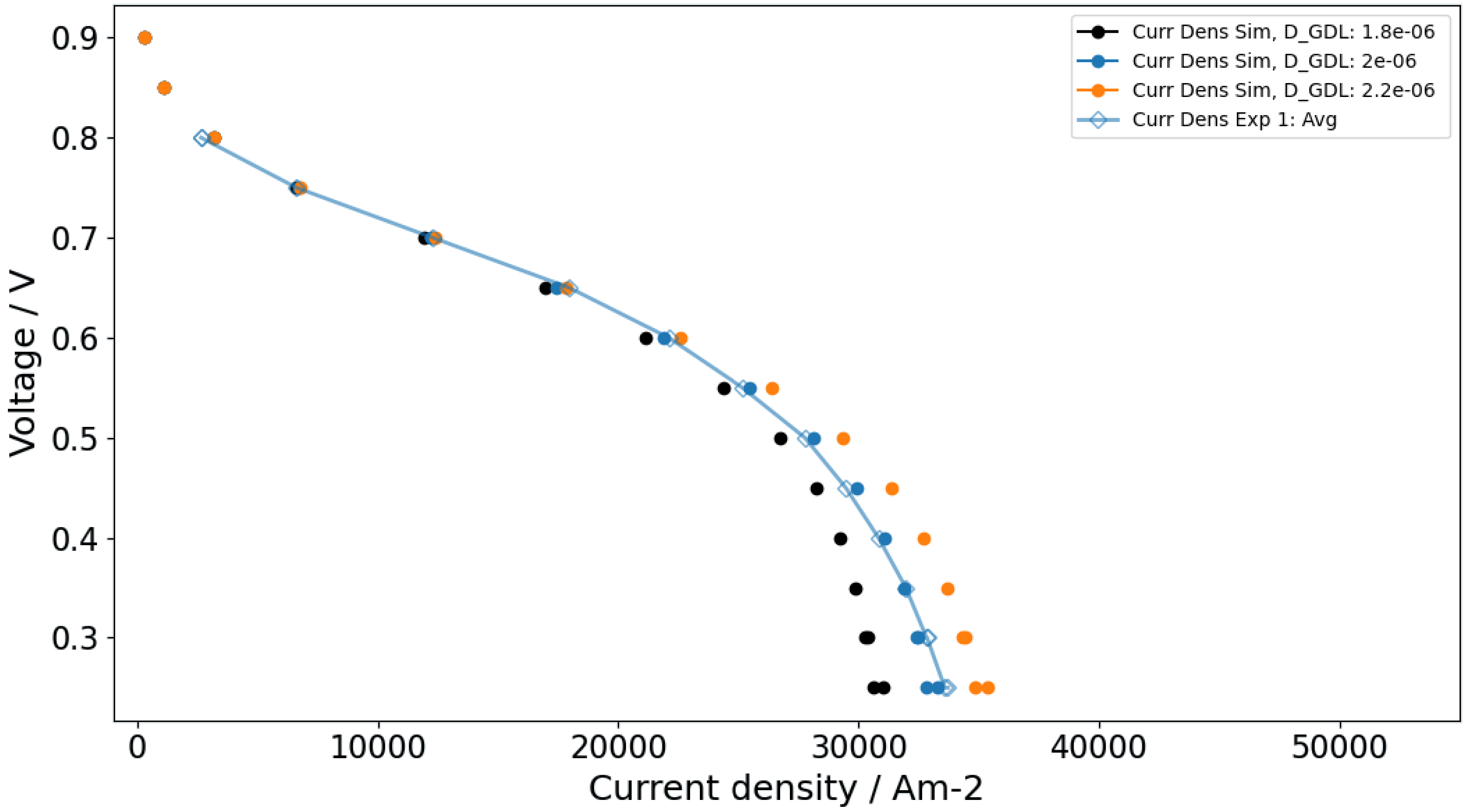
- Polarisation Curve conditions
  - ATC Cell, Counter Flow
  - T_{cool}: 80°C
  - $RH_{a/c}$: 50%
  - $p_{a/c}$: 2 bar abs
  - $\lambda_{a/c}$: 2 / 1.8



Experiment vs ATC Simulation

Polarisation Curve Validation

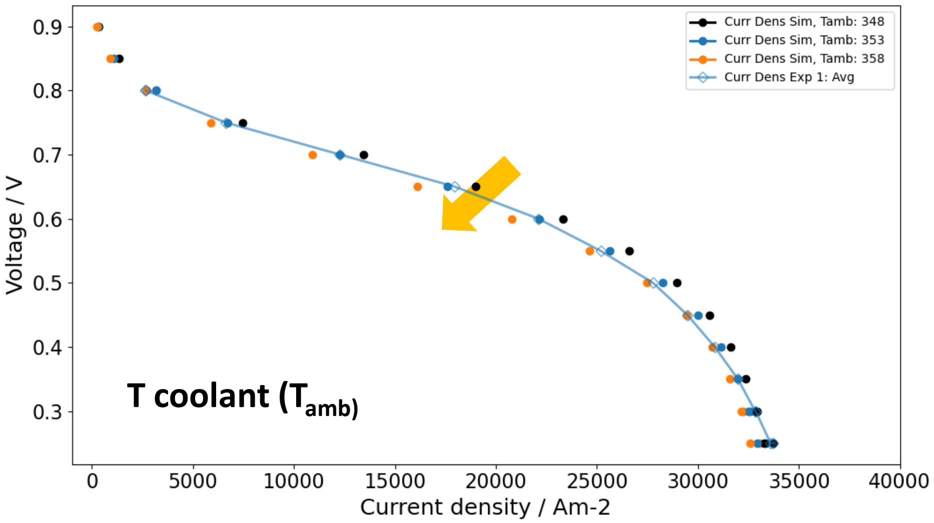
- Polarisation Curve conditions
  - ATC Cell, Counter Flow
  - T_{cool}: 80°C
  - $RH_{a/c}$: 50%
  - $p_{a/c}$: 2 bar abs
  - $\lambda_{a/c}$: 2 / 1.8
- Different Diffusion Coefficients in GDL, good correlation at 2e-6 m²/s

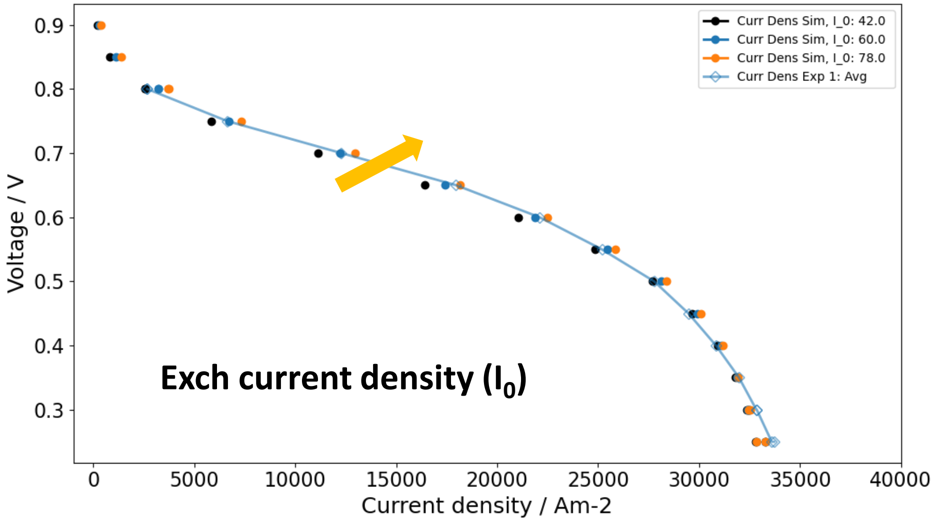


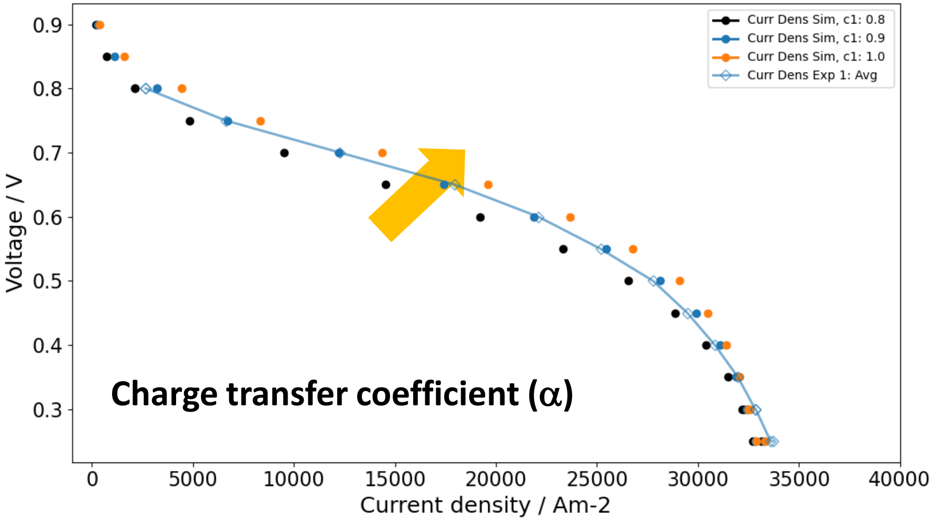
ATC Model

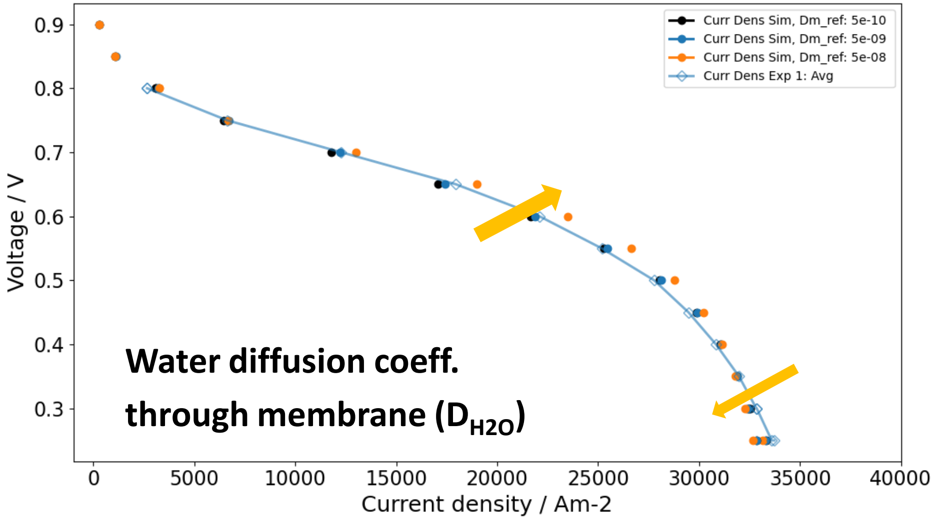
Parameter variation examples

- Many IV-shapes possible with a few parameters
- Physical justification?





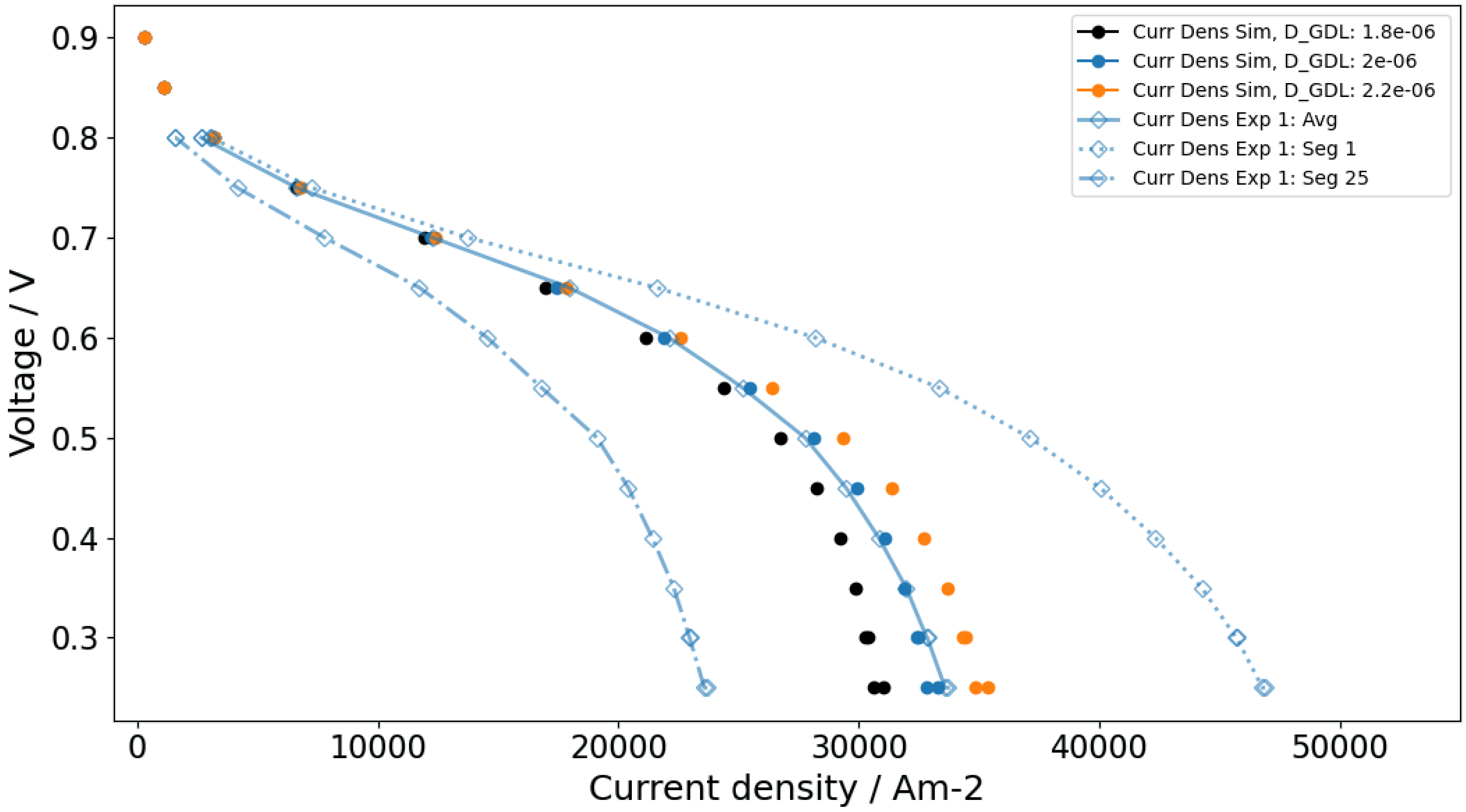




Experiment vs ATC Simulation

Inhomogeneities Along-The-Channel

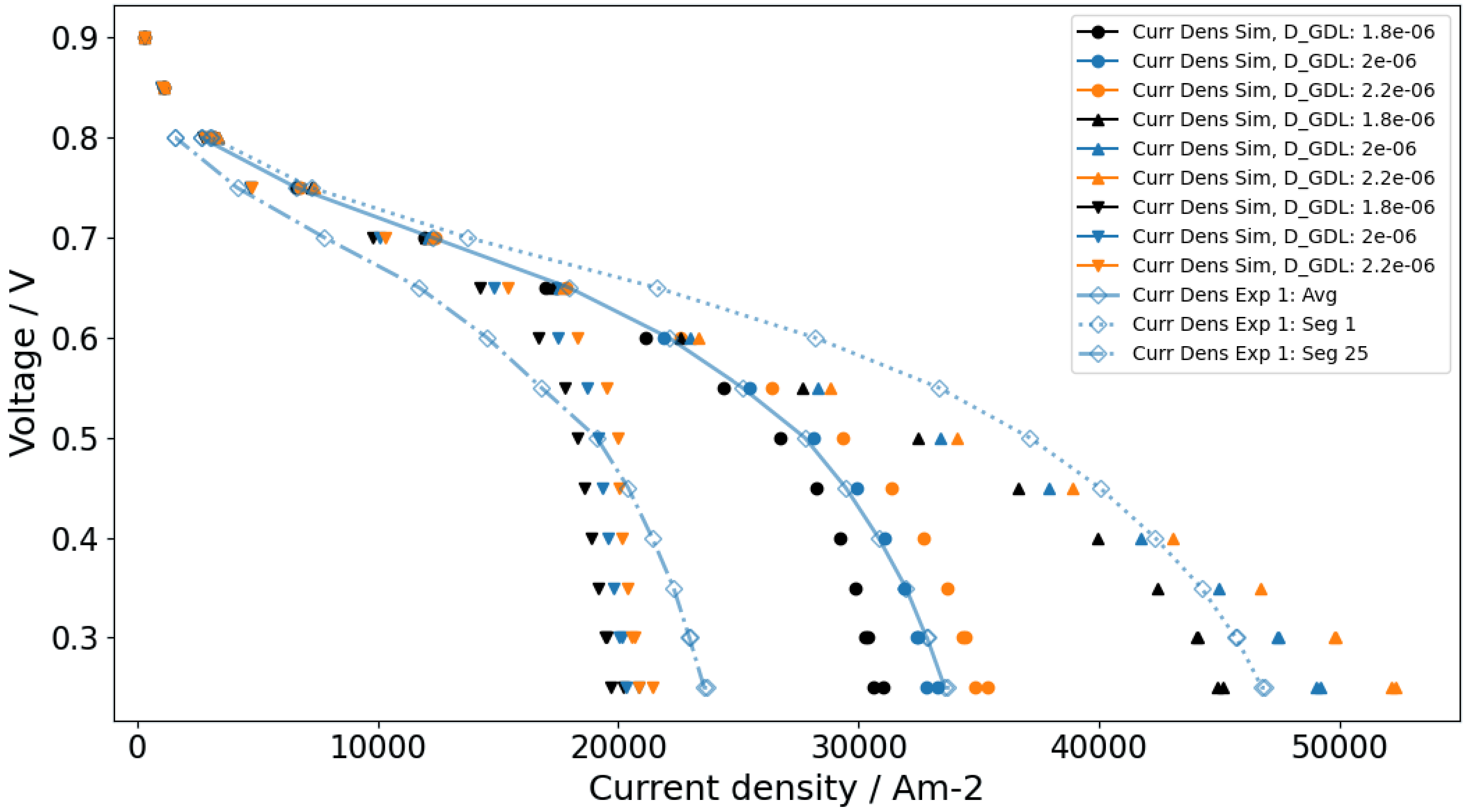
- Large variation (>1 A/cm²) in current density over channel length (25 cm)!



Experiment vs ATC Simulation

Inhomogeneities Along-The-Channel

- Large variation (>1 A/cm²) in current density over channel length (25 cm)!
- ATC model with 25 segments shows same trend for segment 1 and 25, but not exact shapes

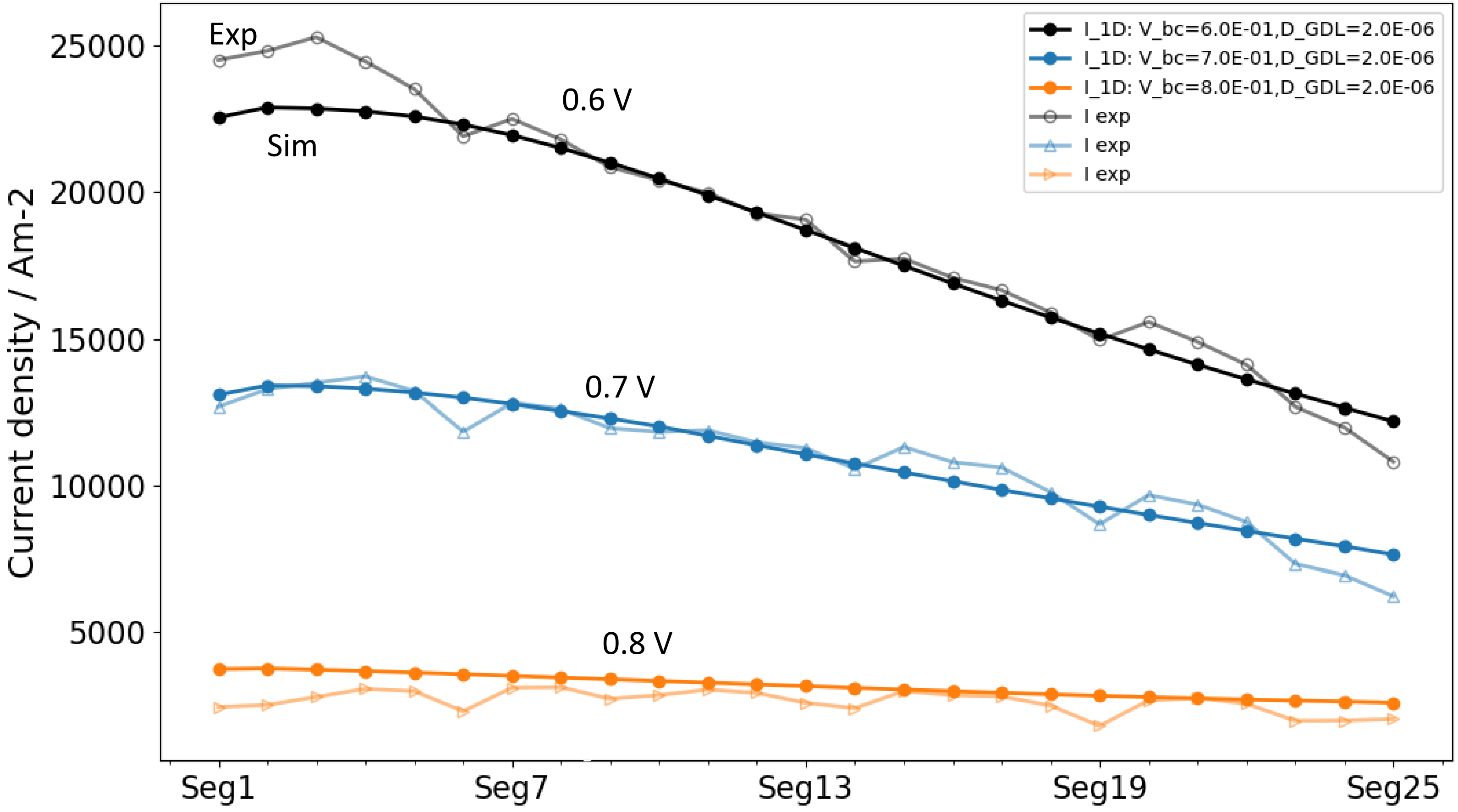


Experiment vs ATC Simulation

Current Distribution Along-the-Channel

- Simulated current distribution after parametrization shows
  - similar shapes
  - correct hight

  for all 3 operating points



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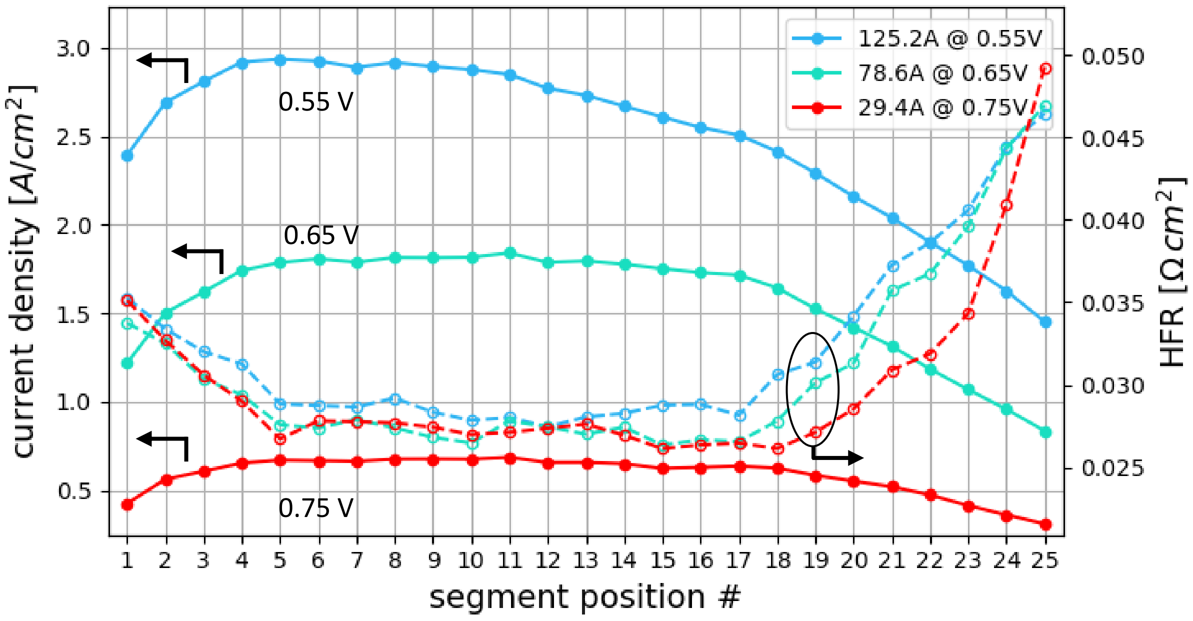
Experiment 2 vs ATC Simulation

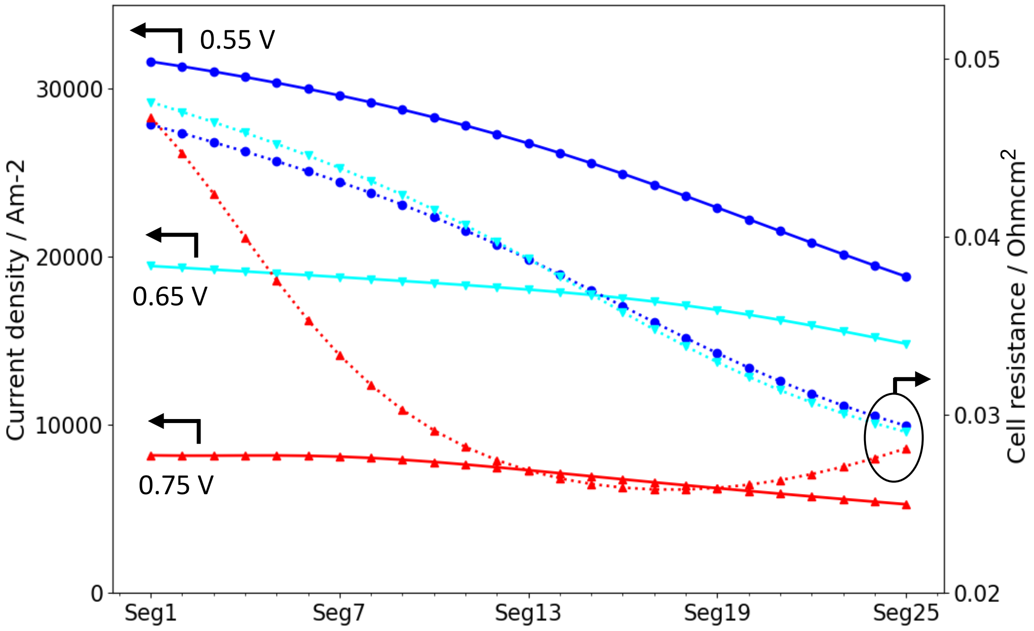
”Bathtub“ HFR shape in experiment

Exp2: I and HFR for 3 voltage points (RH 30/70)



- HFR dominates current distribution

ATC Model: I and HFR

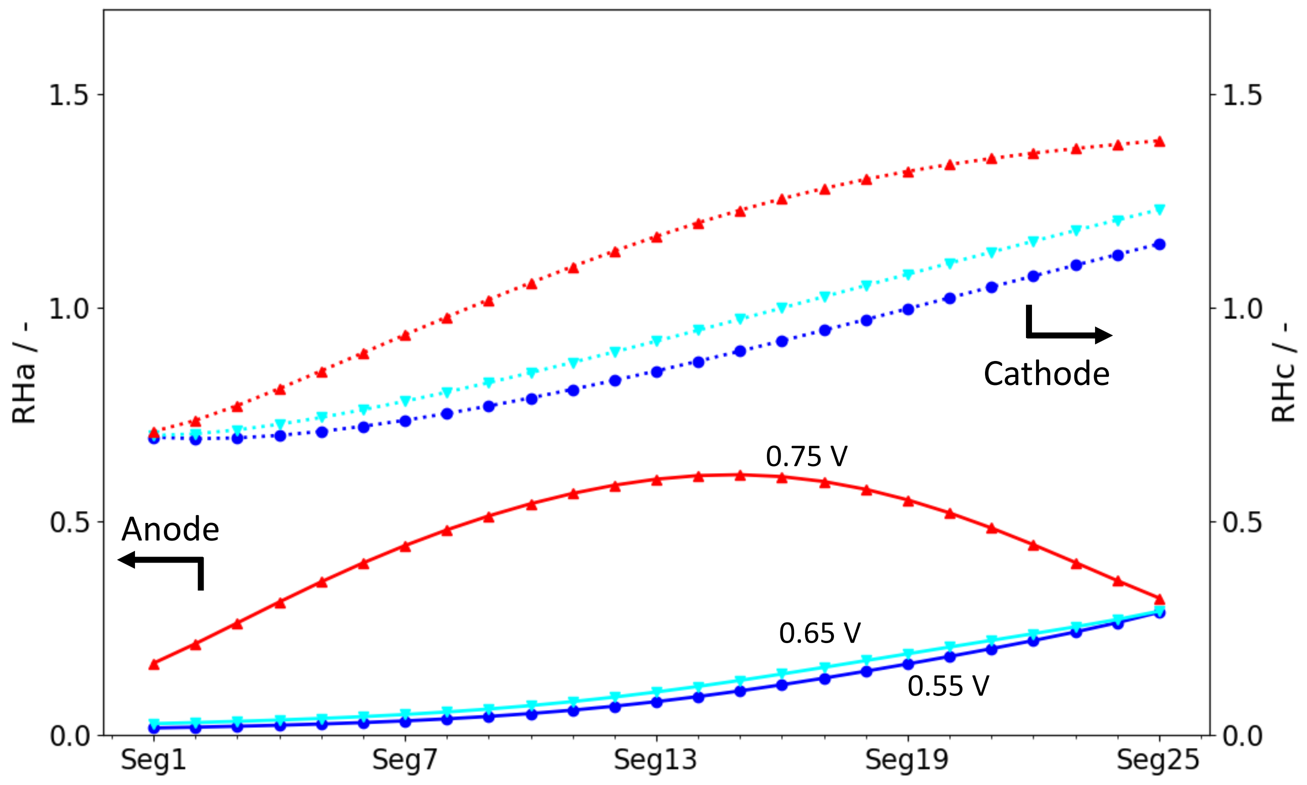


- Similar current ranges and shapes
- HFR ”Bathtub“ only at 0.75 V

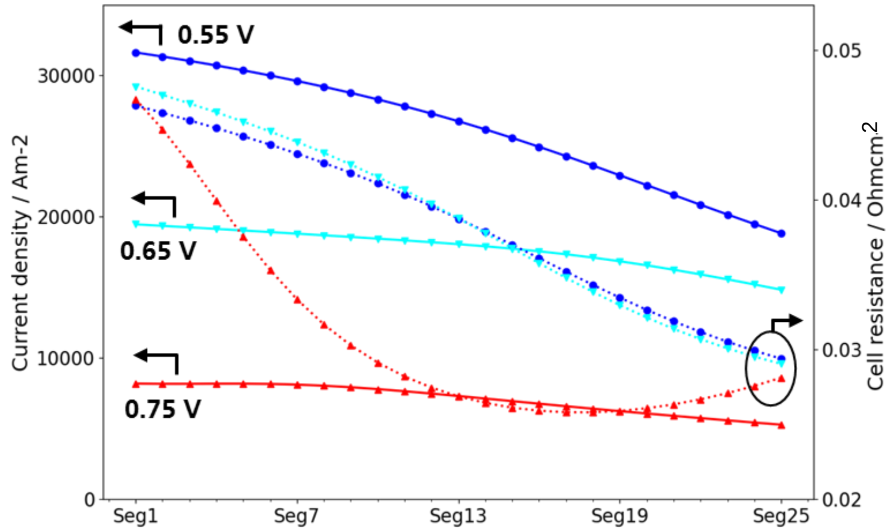
Experiment 2 vs ATC Simulation

Where does the HFR Shape come from?

ATC Model: RH anode and cathode



ATC Model: I and HFR

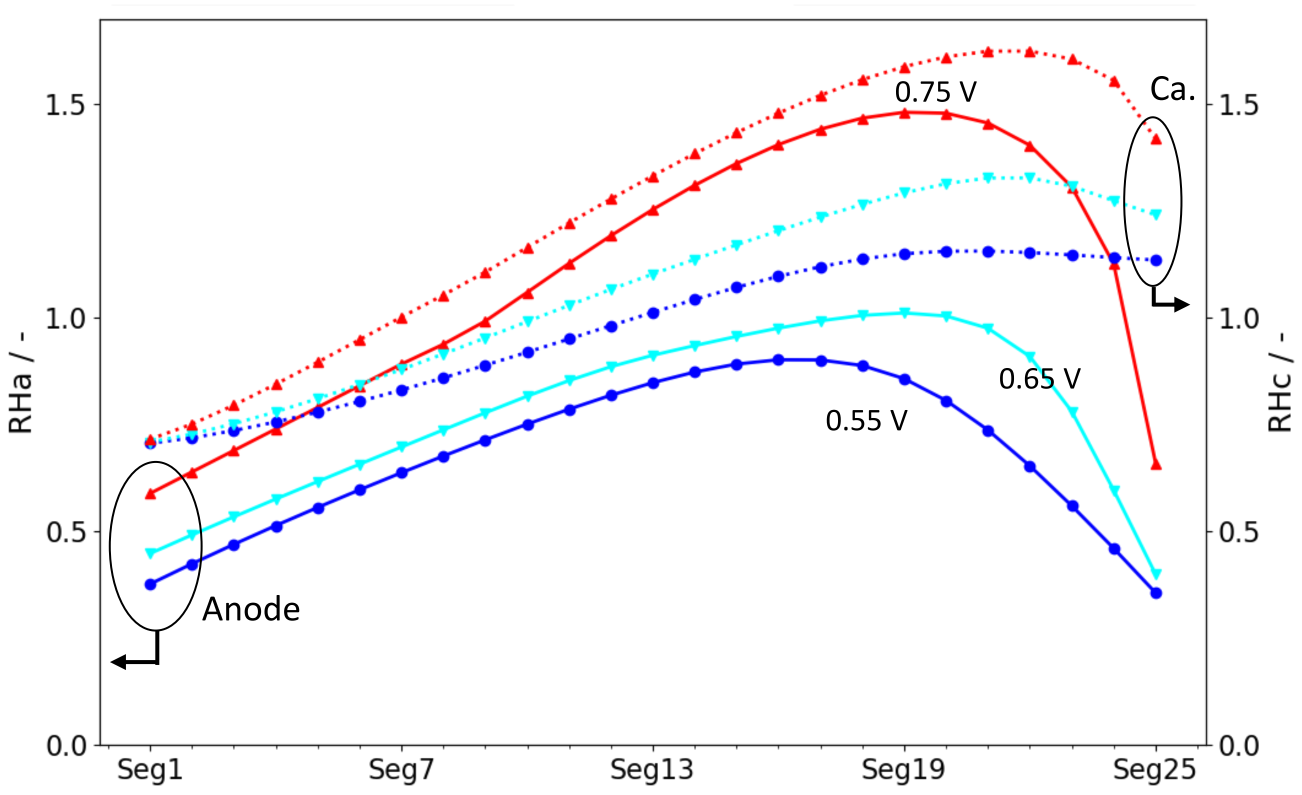


- RHc > 1 near air outlet
- RHa decreases from inlet to outlet (osmotic drag)
- RHa differs at 0.75 V: Opposite curvature
- Lower flow rate at lower current
  → better anode humidification due to longer dwell time
- Adapting H_2O diffusion coefficient through Membrane?

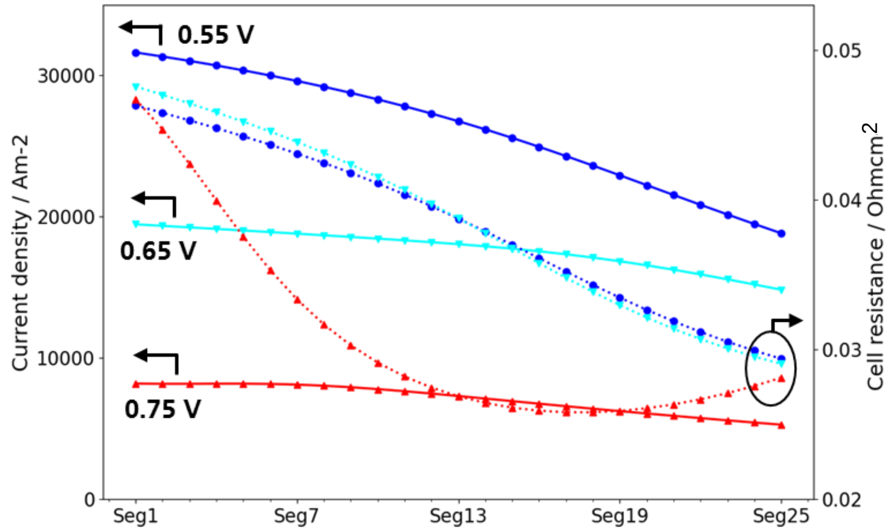
Experiment 2 vs ATC Simulation

RH shapes after parameter change

ATC Model: RH anode and cathode



ATC Model: I and HFR



- RHc > 1 near air outlet
- Lower RHa decrease
- RHa differs at 0.75 V: Opposite curvature
- Lower flow rate at lower current
  → better anode humidification due to longer dwell time
- H2O Diffusion coefficient through Membrane **x 10**
- **HFR: Better "Bathtub" shape but no correct size. Increase on both sides and for all voltages**
- **Current: Better shape at air inlet**

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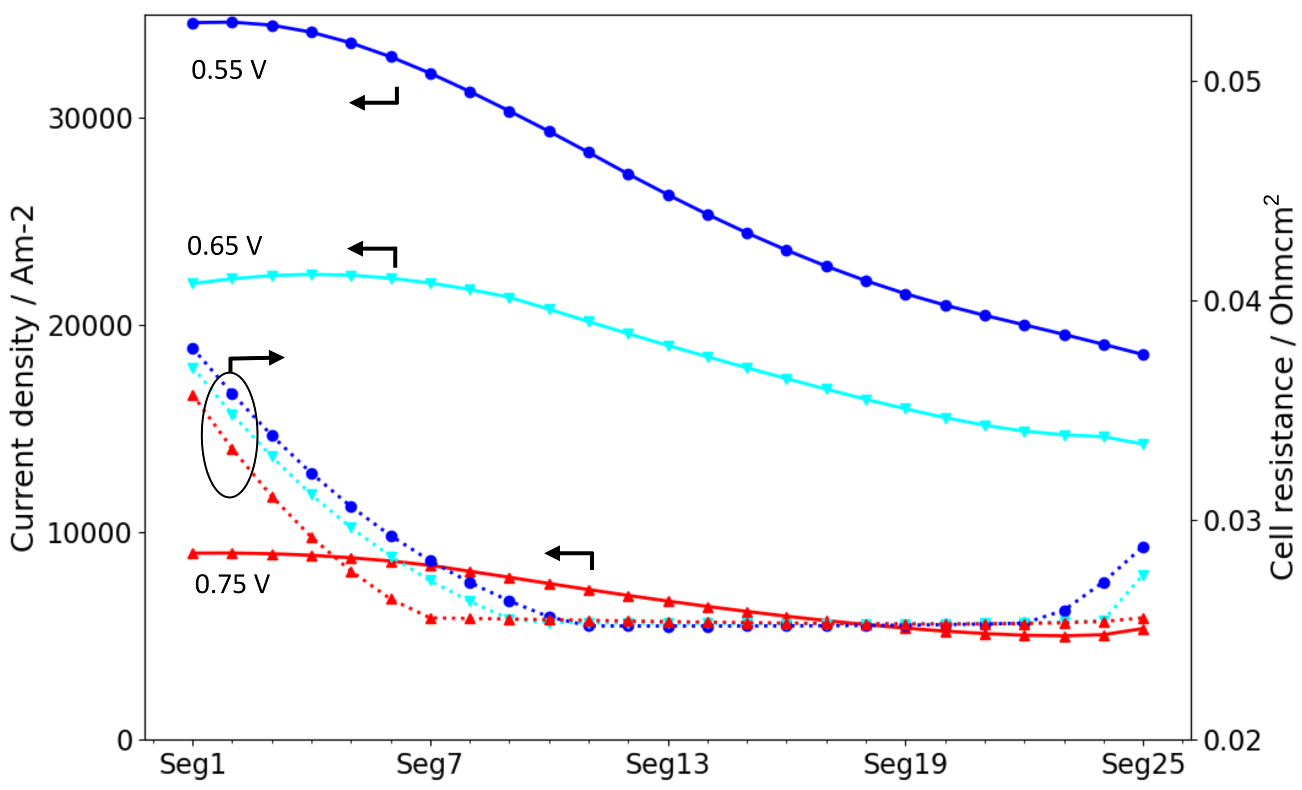
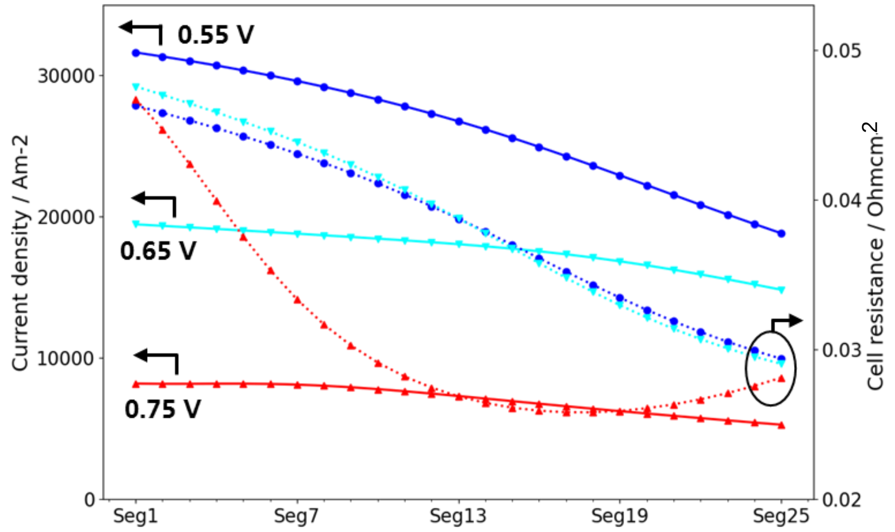
Experiment 2 vs ATC Simulation

Current and HFR shapes after parameter change

ATC Model (changed D_{H2O}): I and HFR



ATC Model: I and HFR



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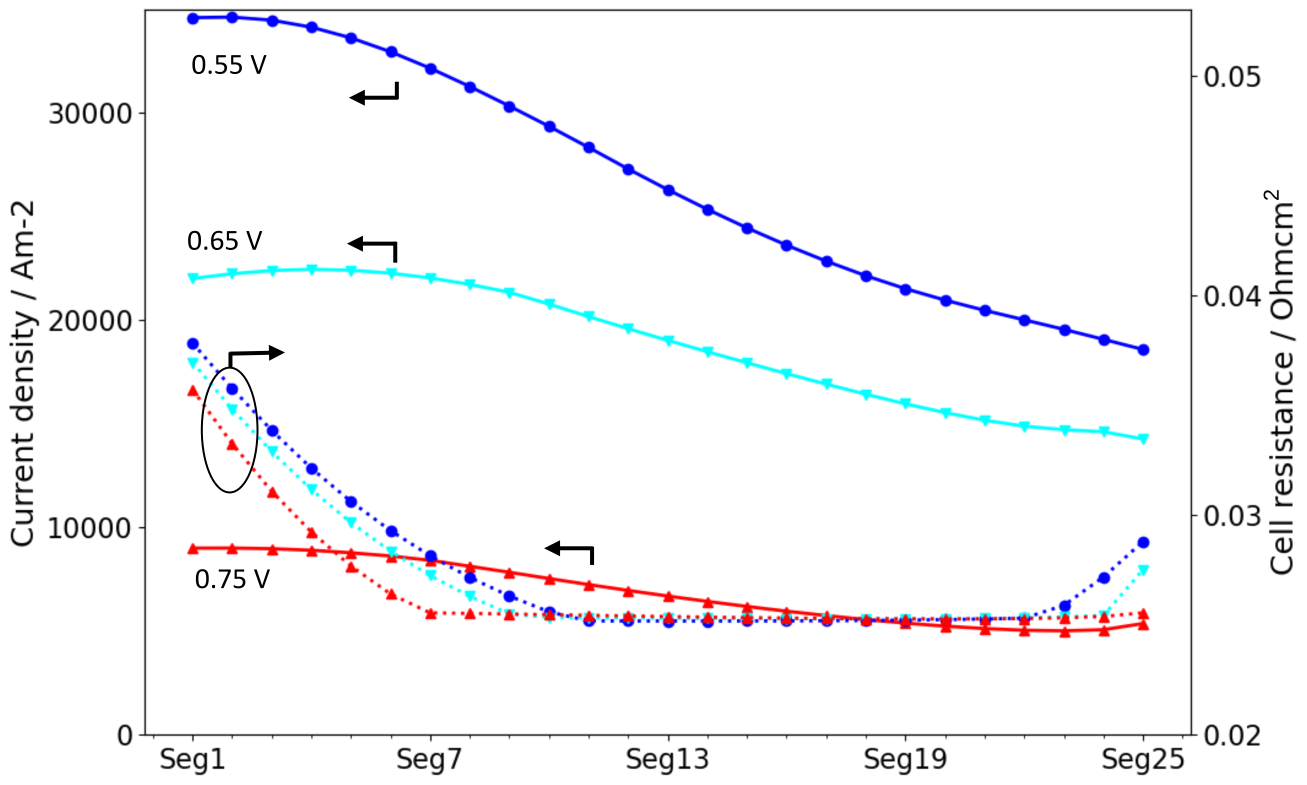
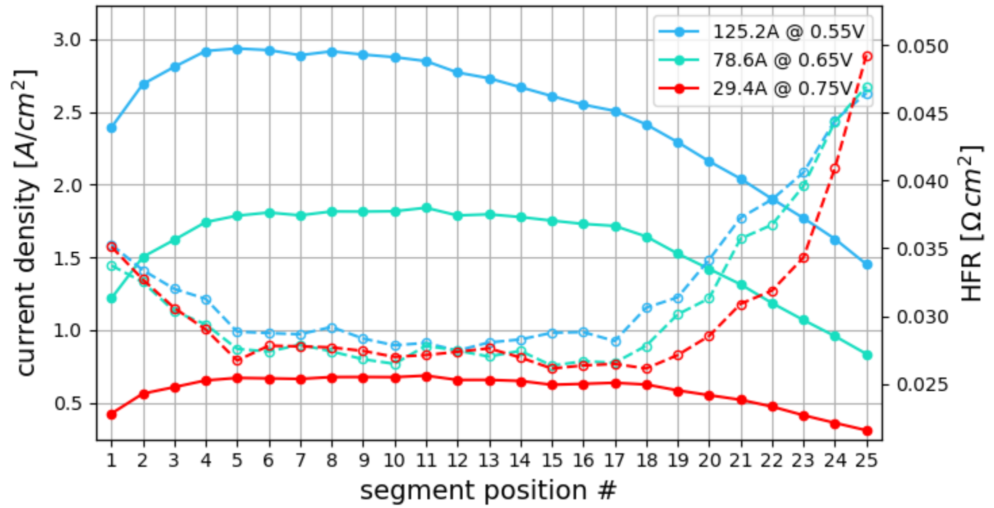


Exp2: I and HFR Distribution



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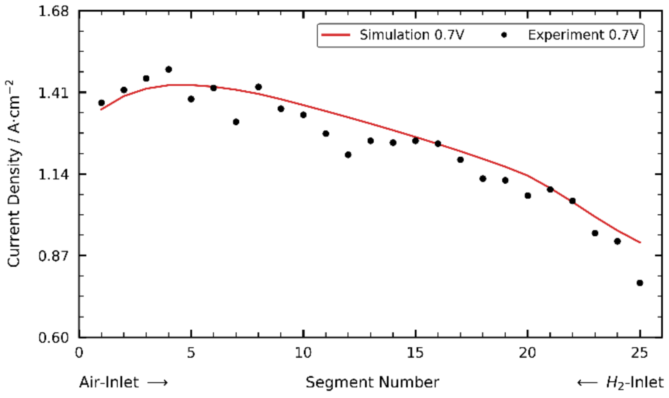
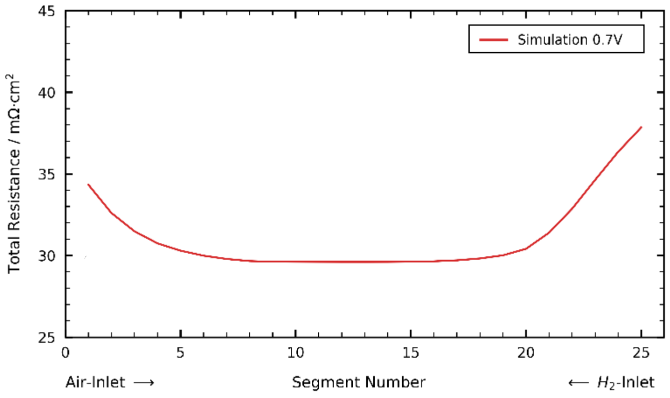
CFD Results - Experiments

Current and HFR distribution at 0.7 V

CFD + Exp1: Current Distribution



CFD: HFR Distribution



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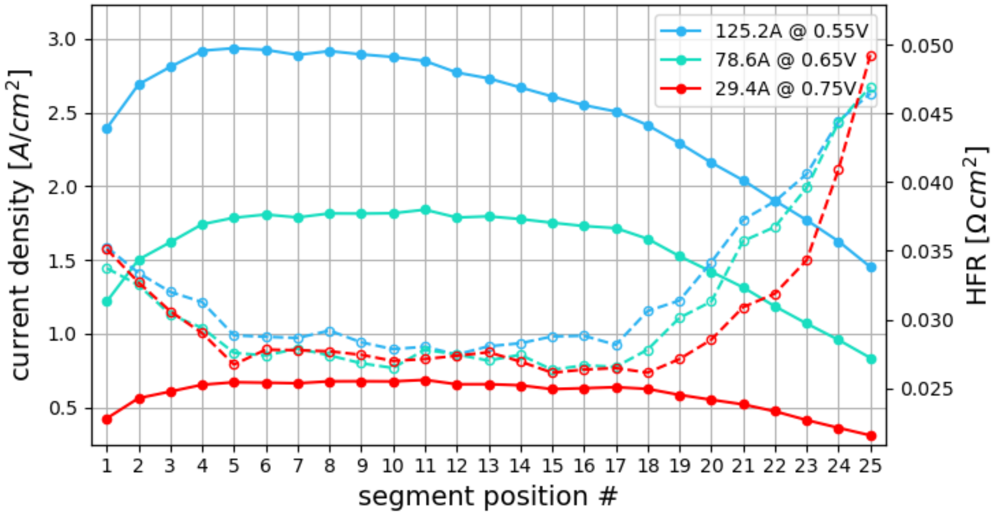
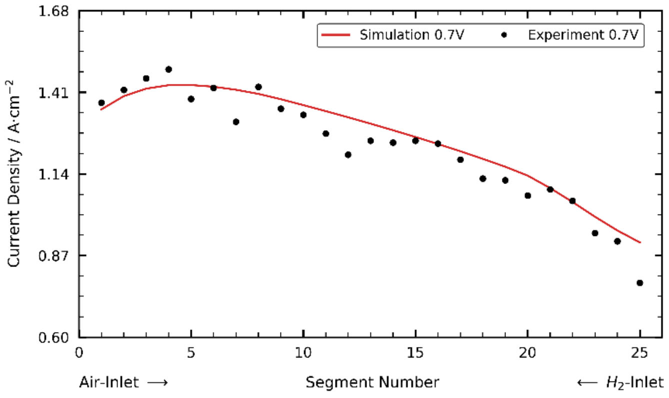


- Current shows detailed local effects
  - Dry air inlet
  - Oxygen depletion
  - Ohmic drop (osm. drag, dry hydrogen)
- Simulated HFR: “bathtub” shape
- Simulation shows nearly exact current / HFR shapes and features of experiment

CFD Results - Experiments

Current and HFR distribution at 0.7 V

CFD + Exp1: Current Distribution



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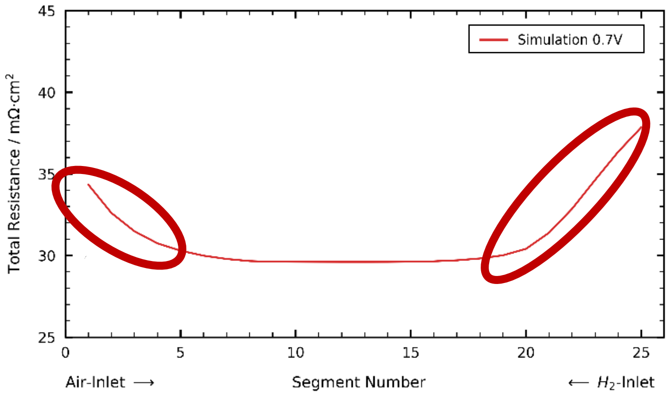

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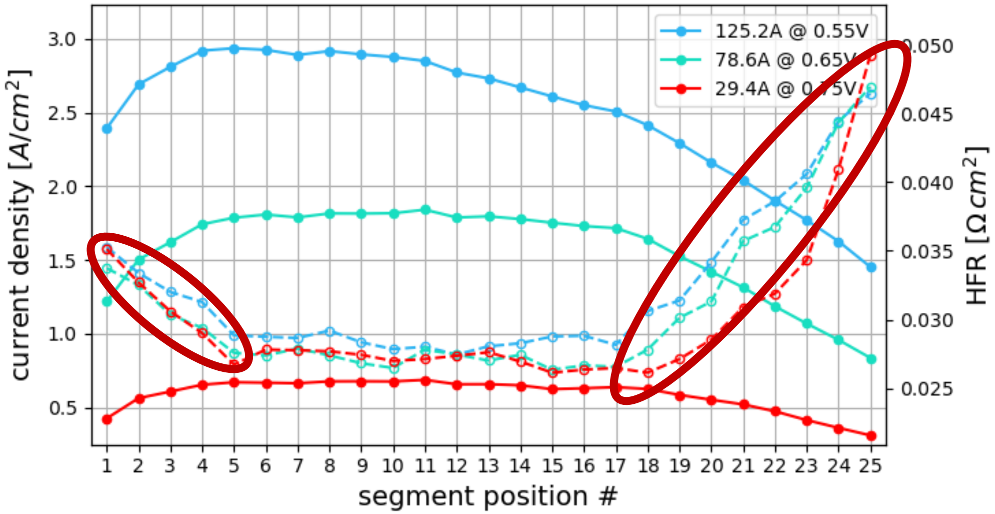

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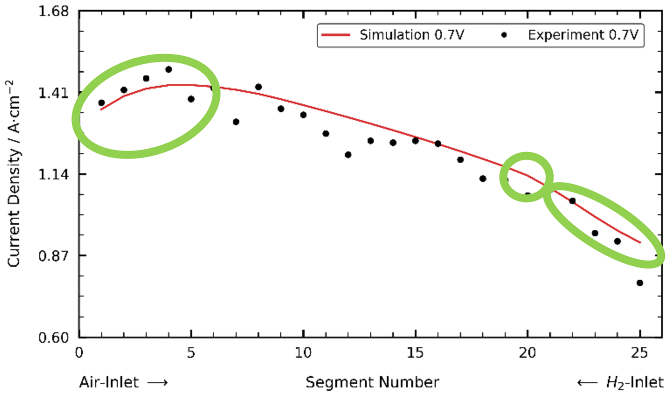

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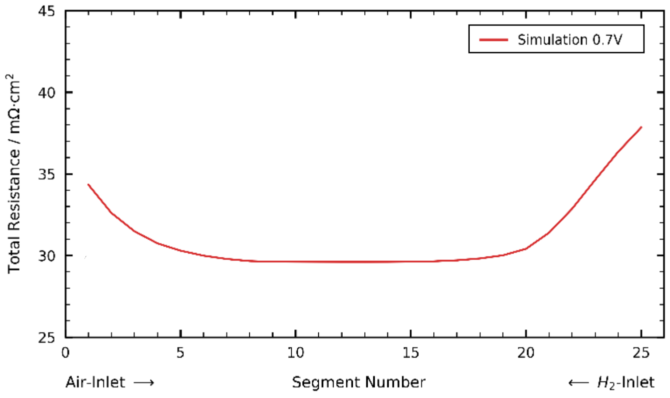

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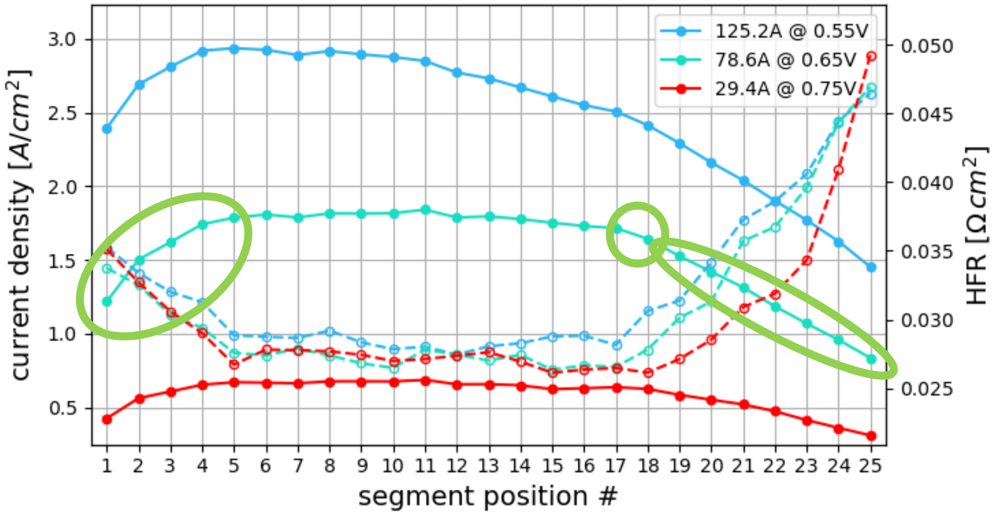
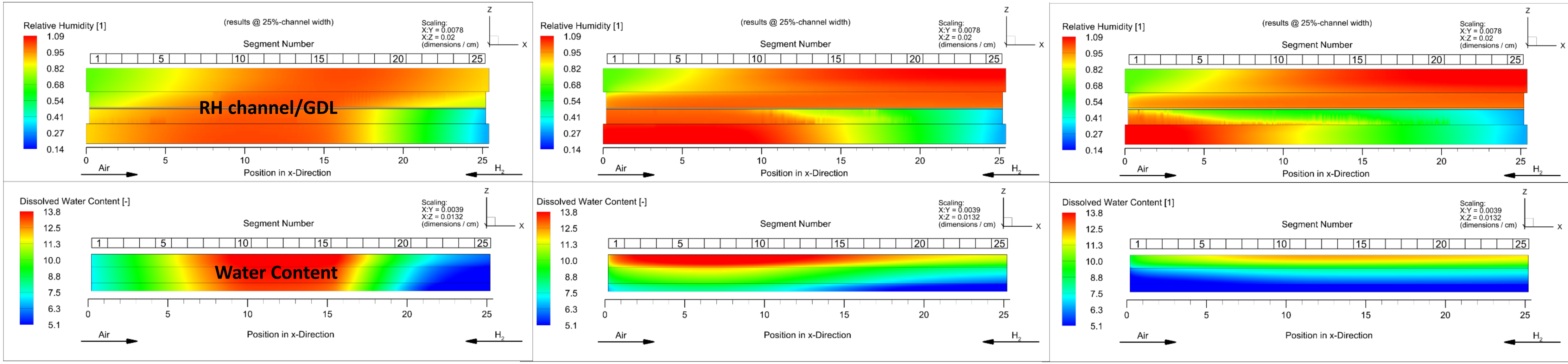


- Simulation shows nearly exact current / HFR shapes and features of experiment

CFD: Humidity distribution in detail: *ATC and TP*

RH in channel/GDL, λ in Ionomer

800 mV, RH 30/70 | 550 mV, RH 30/70 | 300 mV, RH 30/70



- Dry inlet gases on both sides dominate resistance (”bathtub“ shape)
- Small through plane gradients

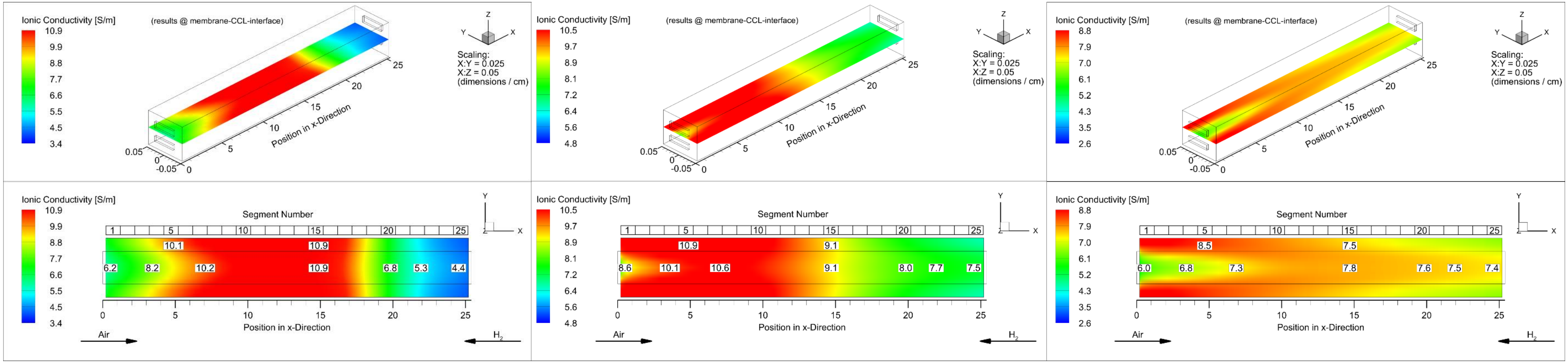
- Higher current production and osmotic drag dominates resistance (flat ”bathtub” / lin. decrease)
- Larger through plane gradient in CCM

- Large through plane gradient in CCM
- Dry anode due to osmotic drag

CFD: Humidity distribution in detail: *Channel - Rib*

Ionic Conductivity (Membrane centre)

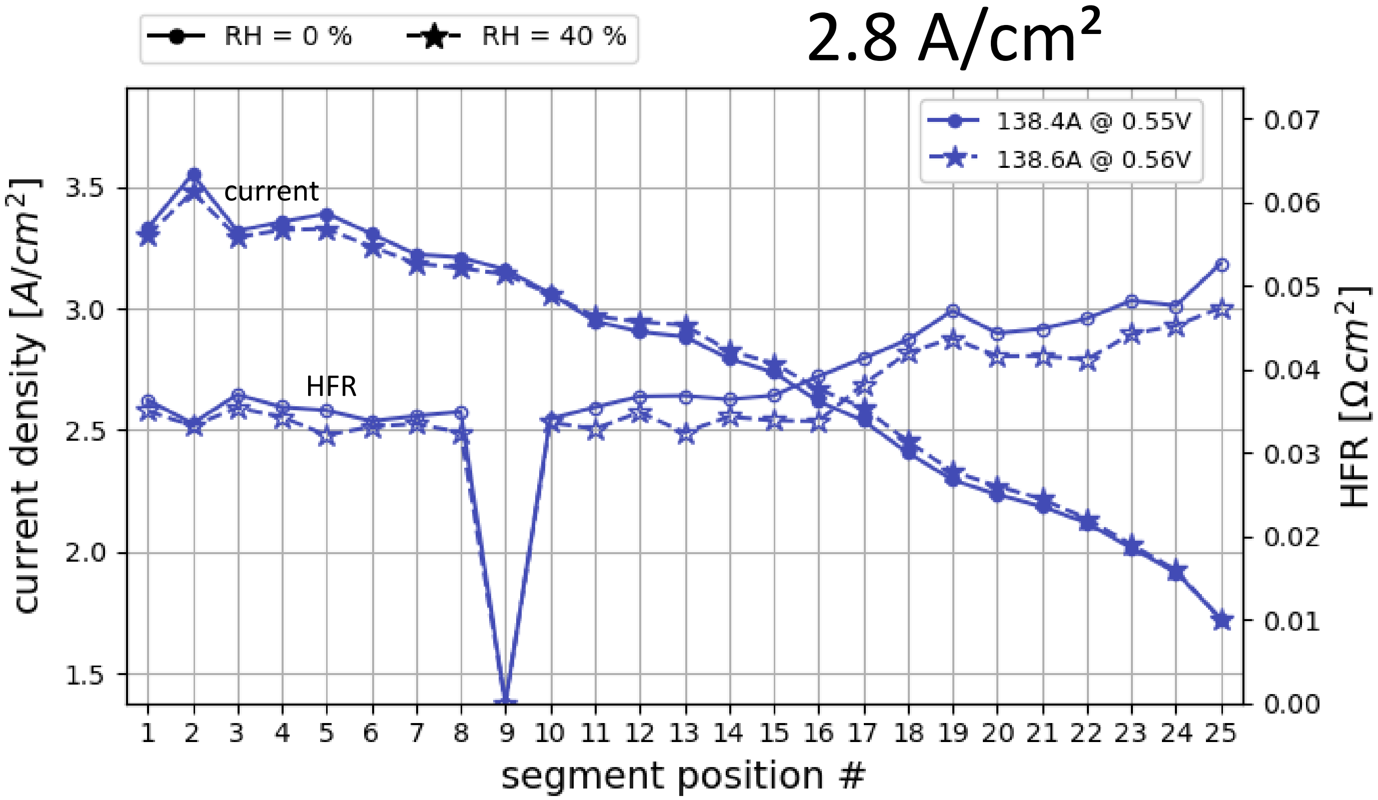
800 mV, RH 30/70 | 550 mV, RH 30/70 | 300 mV, RH 30/70



- Ch-Rib gradients (air inlet) – drying under channel
- Lower Ch-Rib gradients (air inlet) – sufficient water production
- High Gradients – high channel flow , and high Ch-Rib concentration differences

Experiment 3: Drying - RH_c 0% and 40%

Low currents: large difference in current distribution



- **Conditions**
  RHc: 0% / 40%, RHa: 30%, T: 75°C,
  λ_a: 1.4, λ_c: 2.2
- **High current**
  No influence of Air humidity on current density and cell resistance distribution
- **Low current**
  massive decrease in current density at air inlet for dry (RHc = 0) condition
- **@0.8 A/cm² and RHc = 0**
  Completely different current distribution at same total current. First segments are completely dried out → Last segments show higher current

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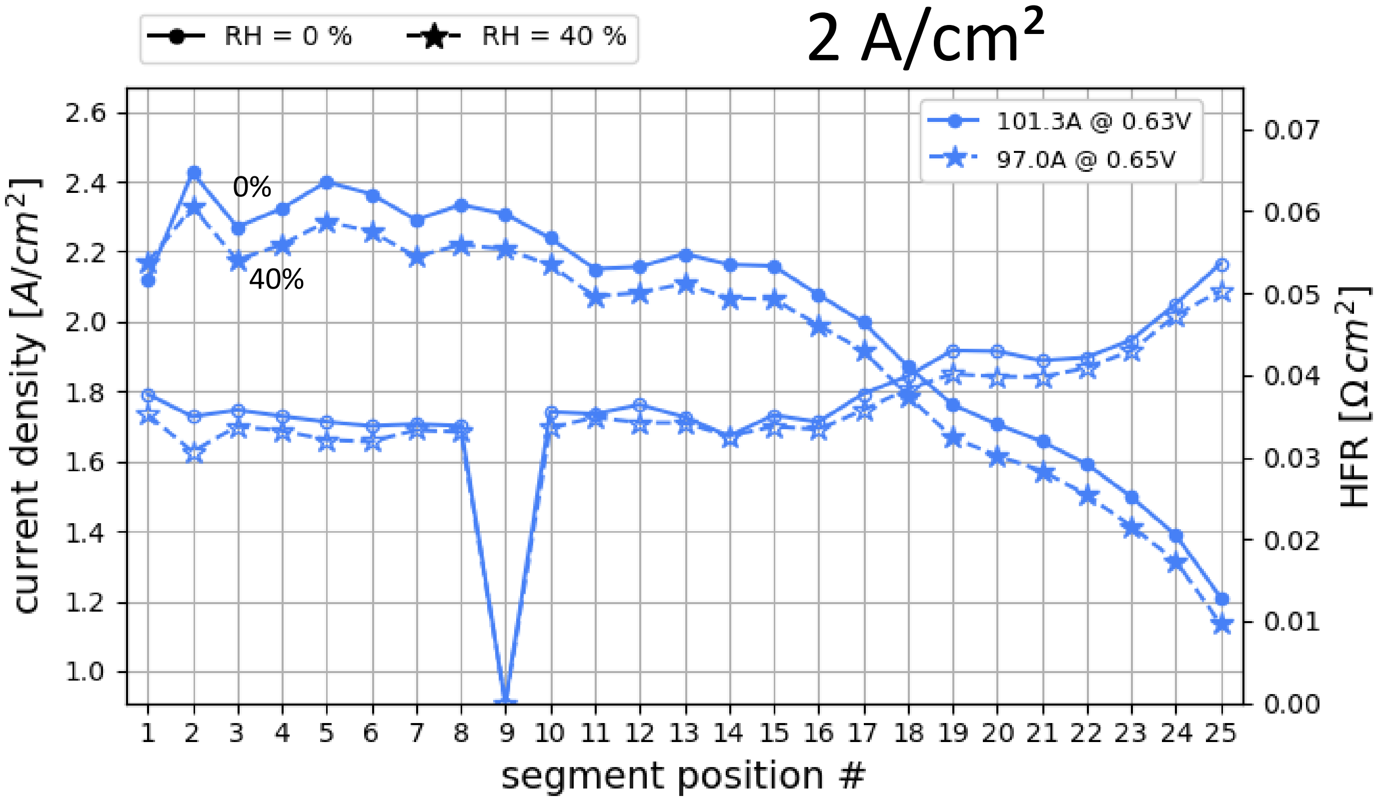

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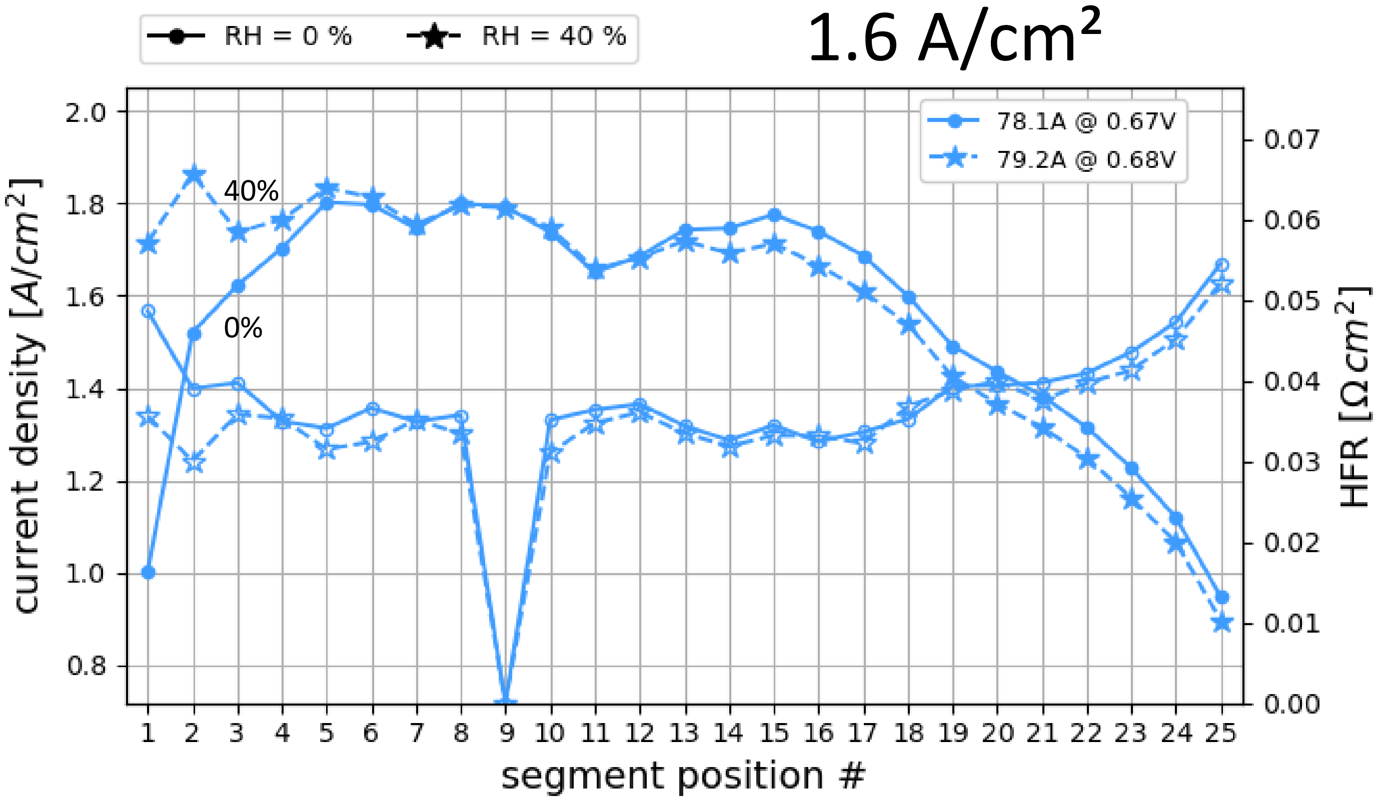

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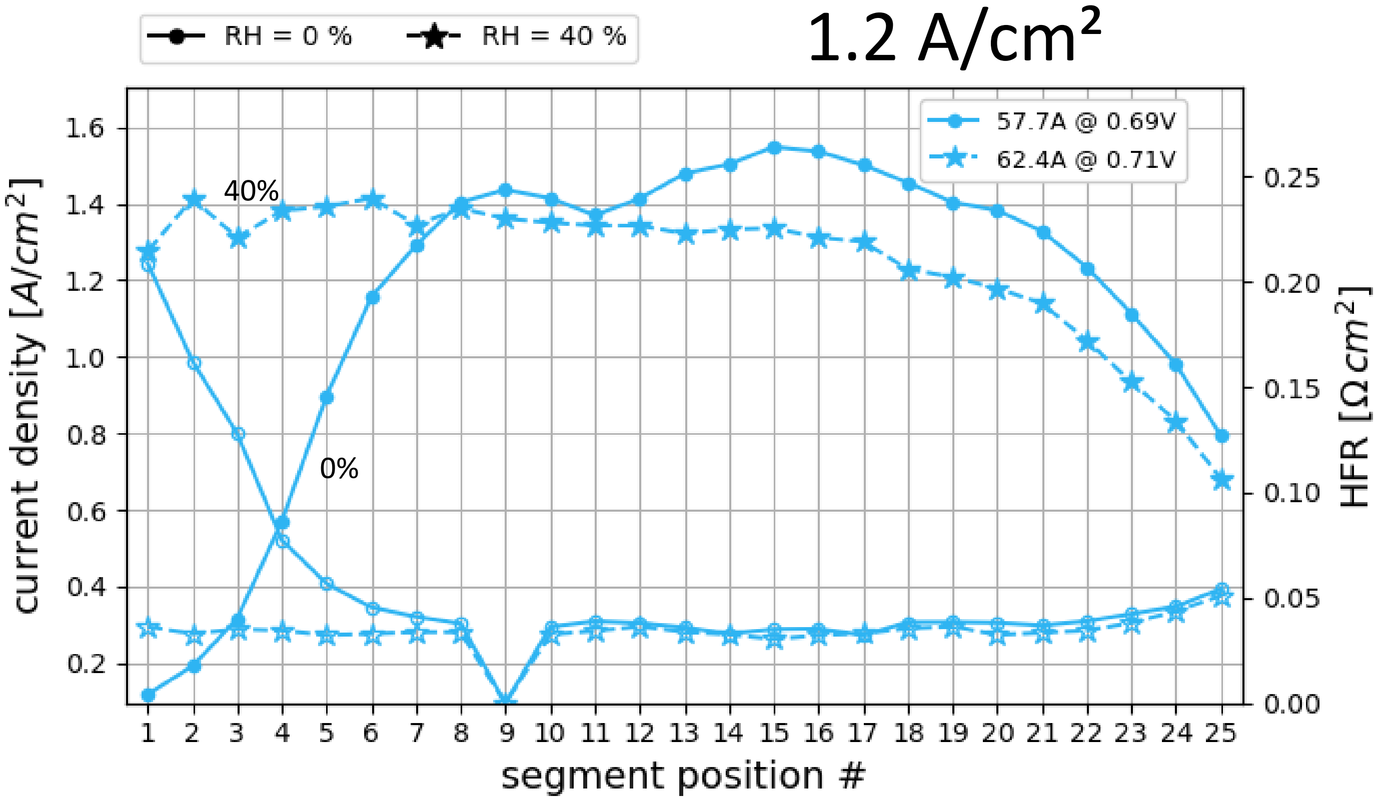

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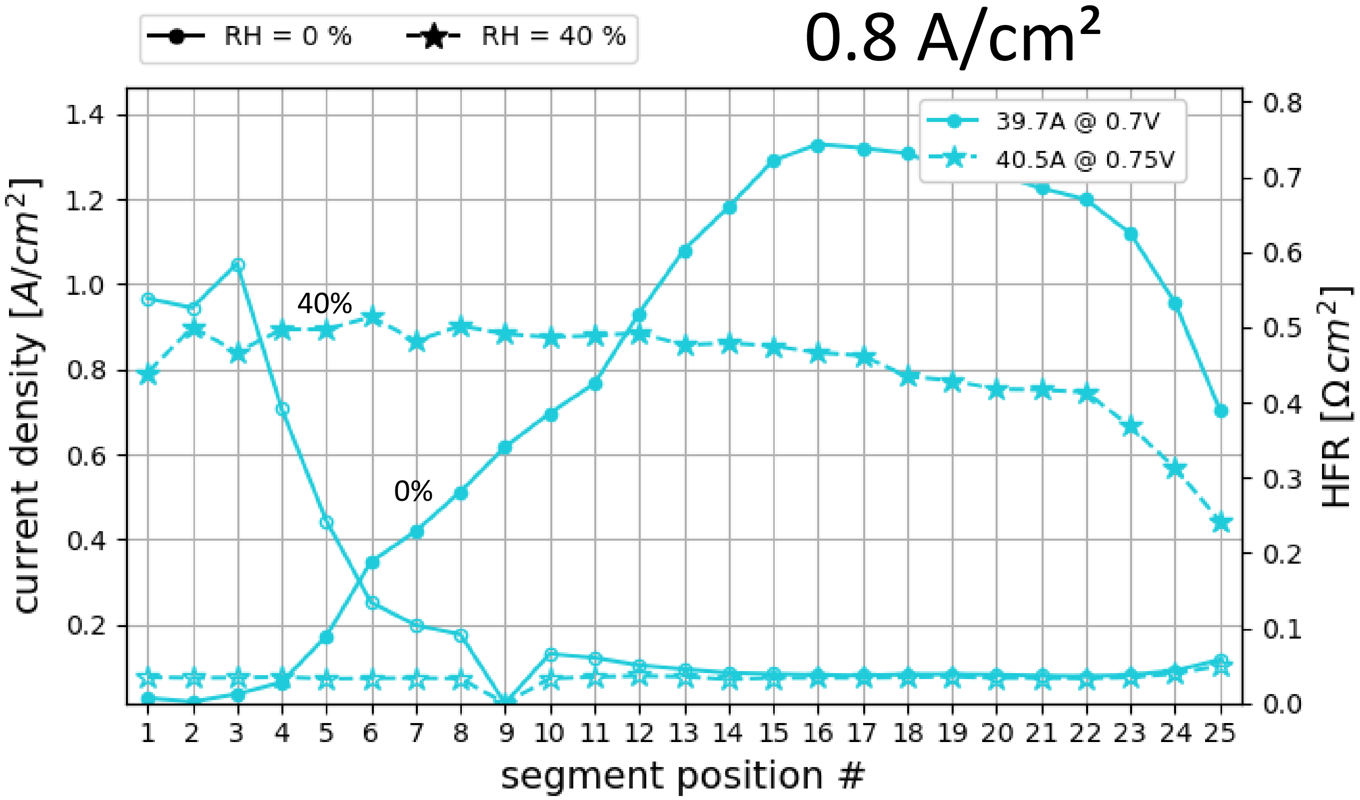
  Completely different current distribution at same total current. First segments are completely dried out → Last segments show higher current

Experiment 3: Drying - RH_c 0% and 40%

Same total current at completely different local current distribution



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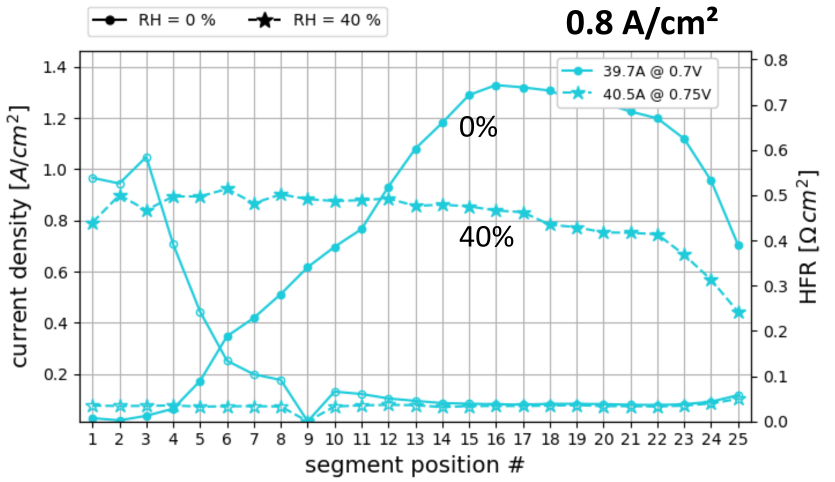
Fraunhofer ISE

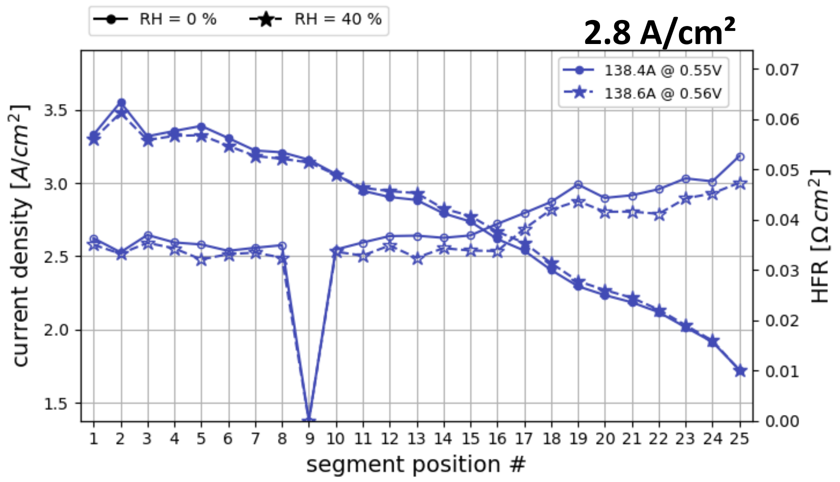
Experiment 3 vs ATC Simulation

Difference at high current

0.8 A/cm²

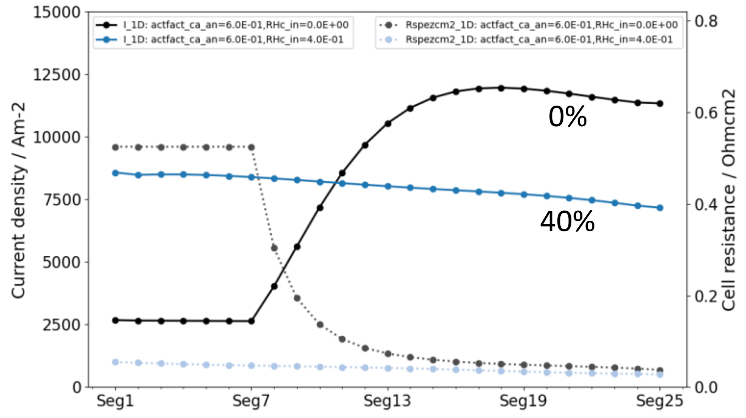


2.8 A/cm²

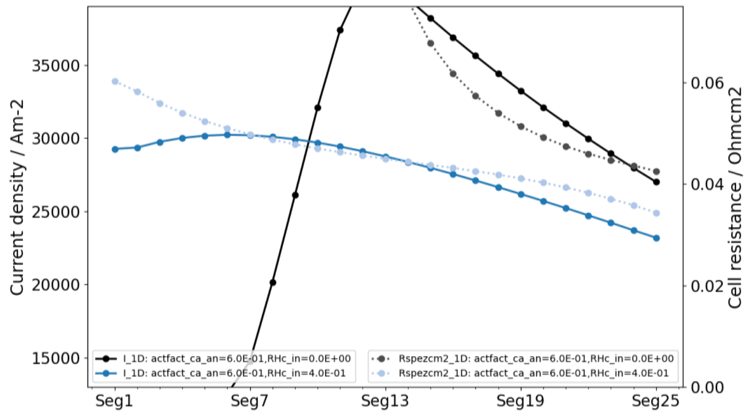


ATC Model

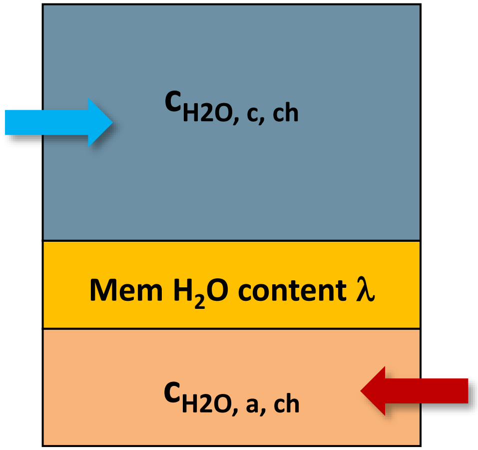
0.8 A/cm²



2.8 A/cm²



- No TP resolution: effect can not be simulated correctly with 1D-ATC model
  → similar behaviour for low and high current

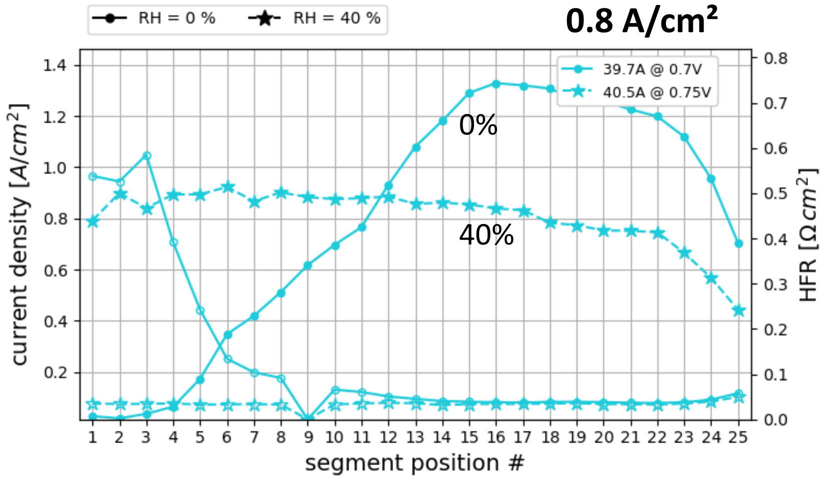


Fraunhofer ISE

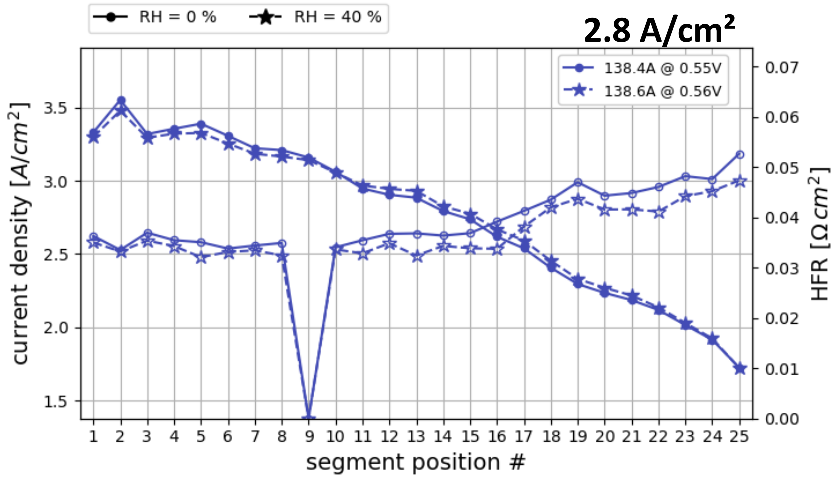
Experiment 3 vs ATC Simulation

Difference at high current

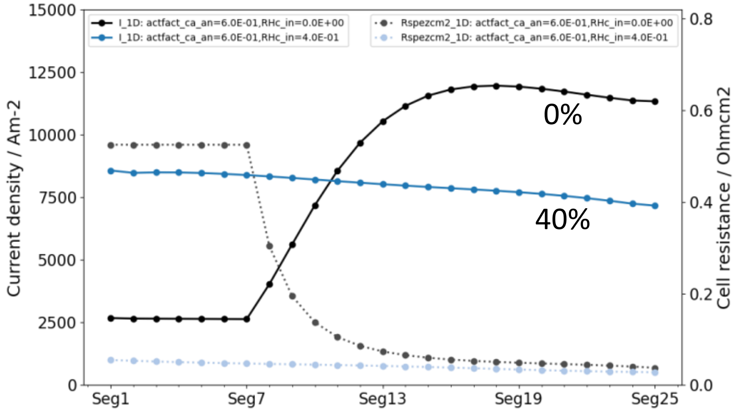
0.8 A/cm²



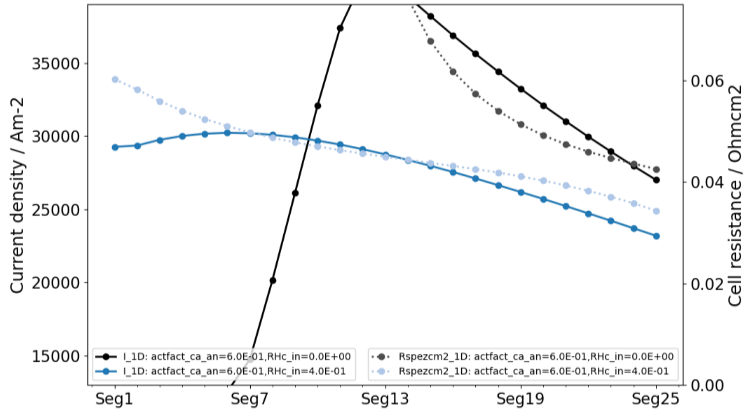
2.8 A/cm²



ATC Model **0.8 A/cm²**



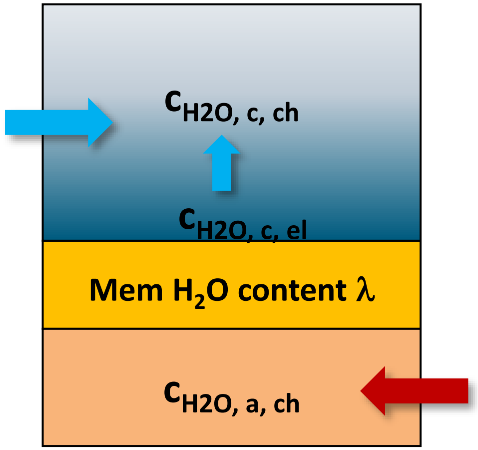
2.8 A/cm²



- New: Through plane gradient in cathode channel H2O concentration (diffusion):

$$c_{H2O,c,el} = c_{H2O,ch} + k \cdot I$$

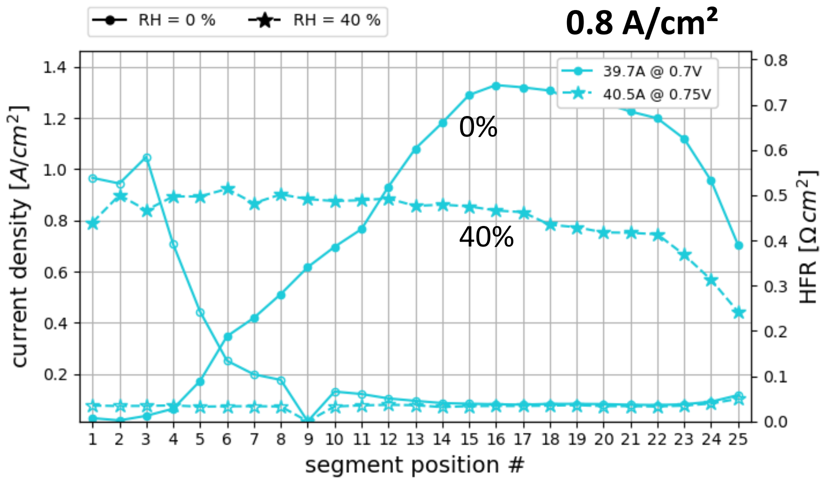
$$\lambda = f(c_{H2O,c,el}, c_{H2O,a,ch})$$



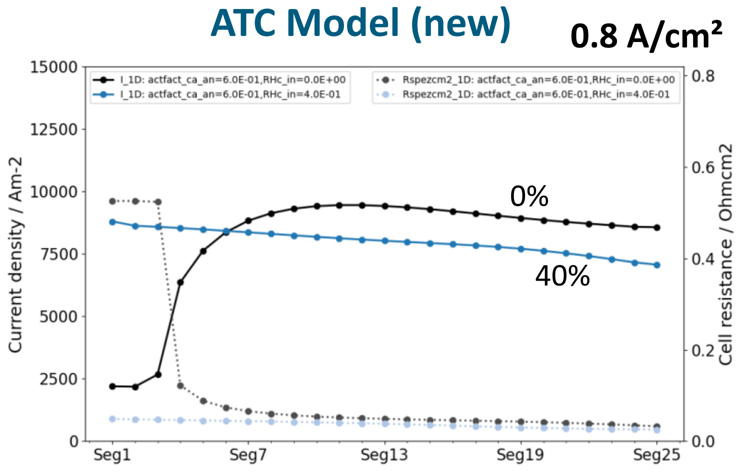
Fraunhofer ISE

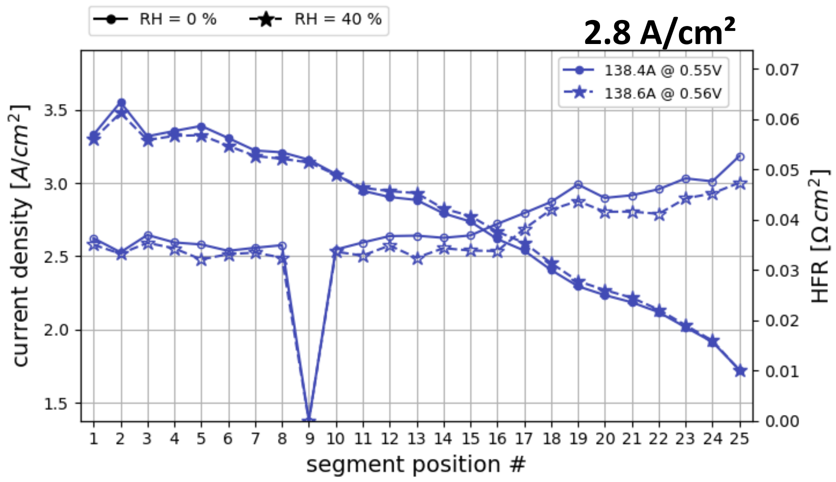
Experiment 3 vs ATC Simulation

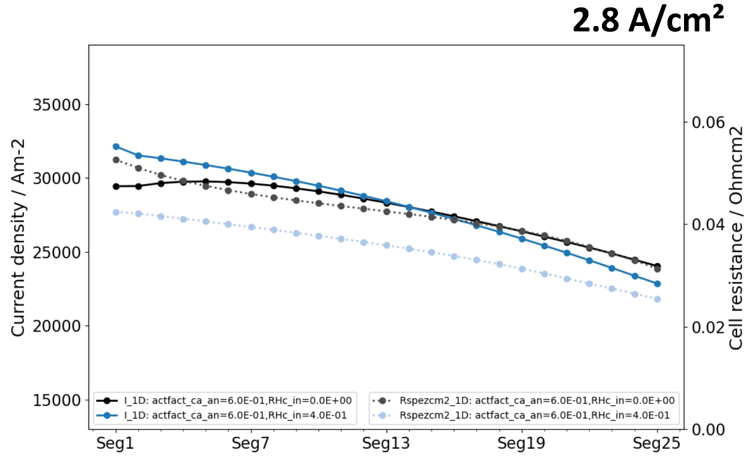
Much better agreement with H2O diffusion in channel



ATC Model (new)







- New: Through plane gradient in cath. channel H2O concentration (diffusion):

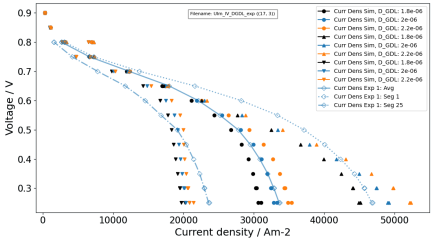
$$c_{H2O,c,el} = c_{H2O,ch} + k \cdot I$$

$$\lambda = f(c_{H2O,c,el}, c_{H2O,a,ch})$$

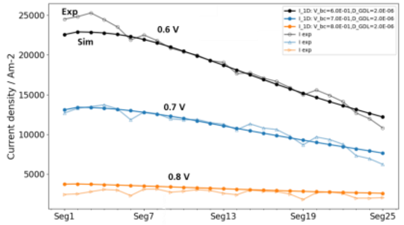
- Much better agreement in all cases. Shapes / features of current distribution are correctly reproduced for high and low current and humidity.
- Membrane resist. shows similar size but wrong tendency at high current

Conclusions

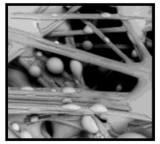
- ATC Model can be a useful tool for interpretation of PEMFC measurements
  - High speed, high flexibility → simulation parallel to experiment
  - Shows variations that are not visible in 0D Model or (ATC) test cell experiments
  - ATC experimental results are helpful for proper model validation

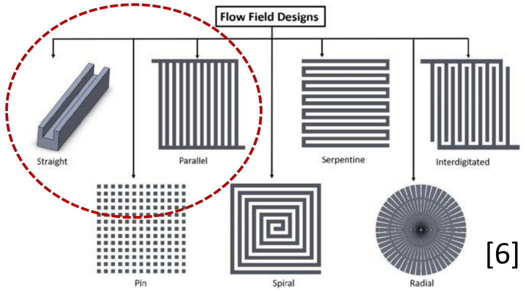


- ATC Model shows good agreement in
  - Steady state performance (IV-Curves)
  - Current distribution along-the-channel at different op. points (total numbers and curve shapes)
  - Resistance distribution can be reproduced with model adjustments

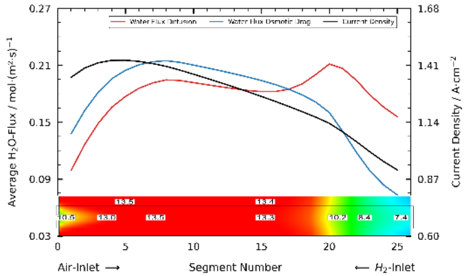


- Limitations
  - Extreme conditions (flooding / drying)
  - Non-parallel channel geometry
  - Conditions with high gradients in through-plane or channel-rib direction
    - More complex (CFD) models preferred





[6]



[6] M. Kumar et al, Performance Studies of Proton Exchange Membrane Fuel Cells with Different Flow Field Designs - Review. Chemical record (New York, N.Y.). 21., 2021

Thank you for your attention

Contact

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