EIS and Impedance Model Background Information

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Finding Me

- LinkedIn
  - https://www.linkedin.com/in/nick-gould/
- Github
  - https://github.com/nick-gould

Ohm’s Law

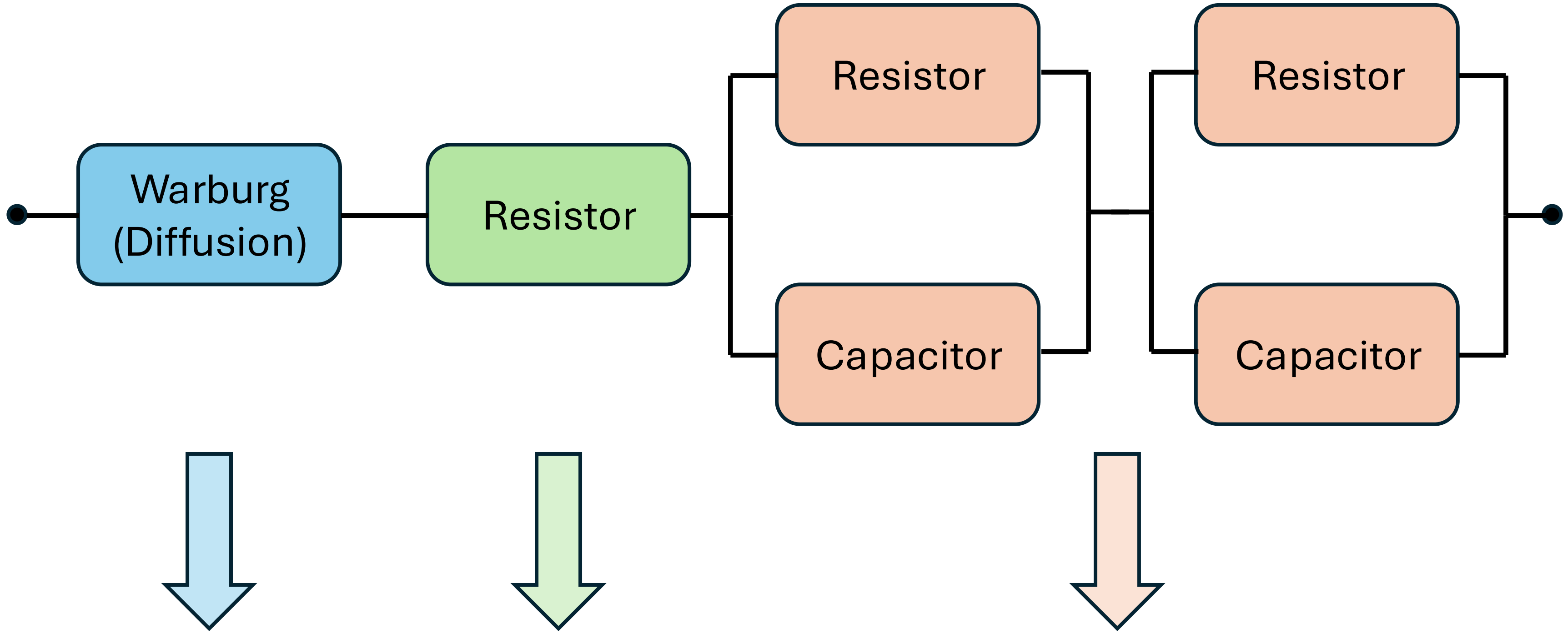
- Battery chemistry is applying Ohm’s Law over and over, where the value for resistance is determined by the build of the cell
- Ohm’s Law can be written as:
    - Change in Voltage = (Change in Current) x Resistance

EIS Background

- Please refer to this reference from Gamry:
  - https://www.gamry.com/application-notes/EIS/basics-of-electrochemical-impedance-spectroscopy/

Impedance Model

- Electrochemical Impedance Spectroscopy (EIS) is a technique for breaking apart a cell into its component parts to determine the resistance contributions of the electrodes, the electrolyte, any interfaces, and mass transfer of the components moving around within the cell
- EIS does this by converting from the time domain to the frequency domain using a Fourier Transform
- EIS requires specific hardware, special handling of the data, and a lot of time
    - EIS-capable testing channels cost more normal testing channels
    - EIS happens in the frequency domain, requiring separate files from typical time domain charging and discharging
    - If low frequency measurements are needed, it can take upwards of 24 hours for a single data point



$$Z(s) = \frac{W}{\sqrt{s}}$$

$$Z(s) = R$$

$$Z(s) = \frac{R}{RCs + 1} \; (each)$$

$$I(s) = \frac{I}{s}$$

For current step

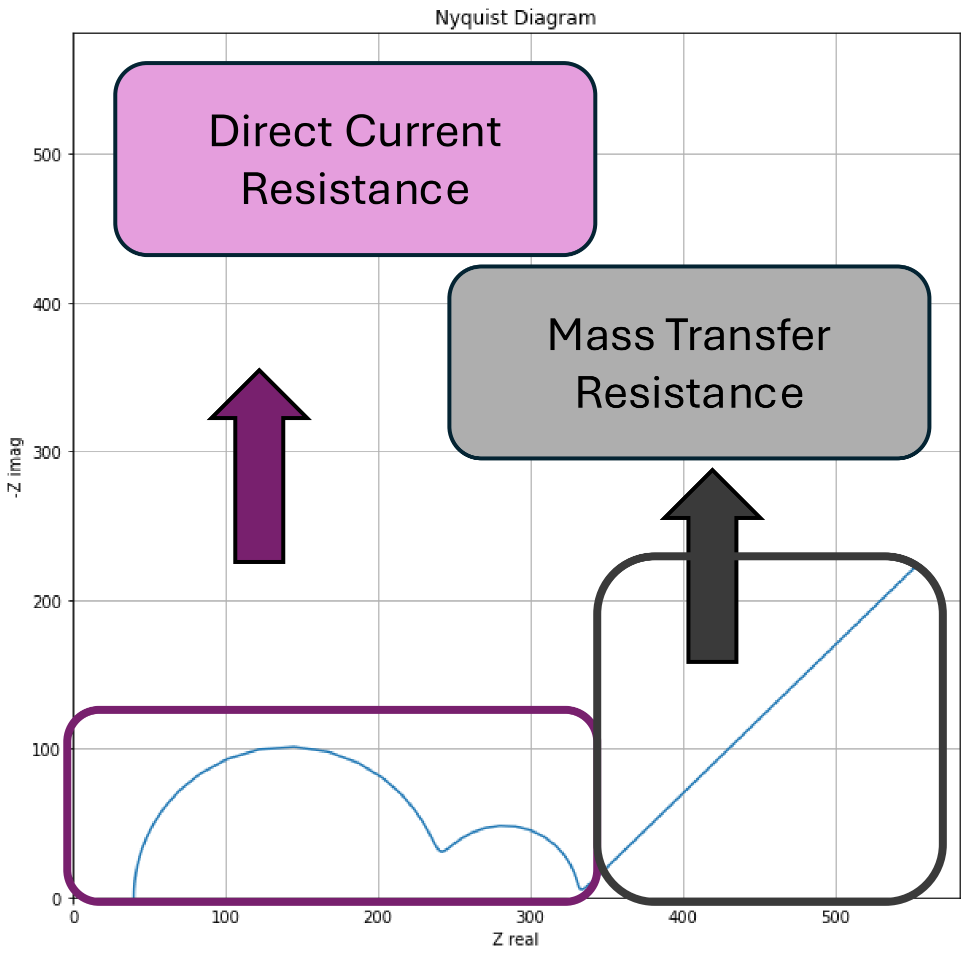
$$Z(s) = \frac{R}{RAs^{\alpha} + 1}$$

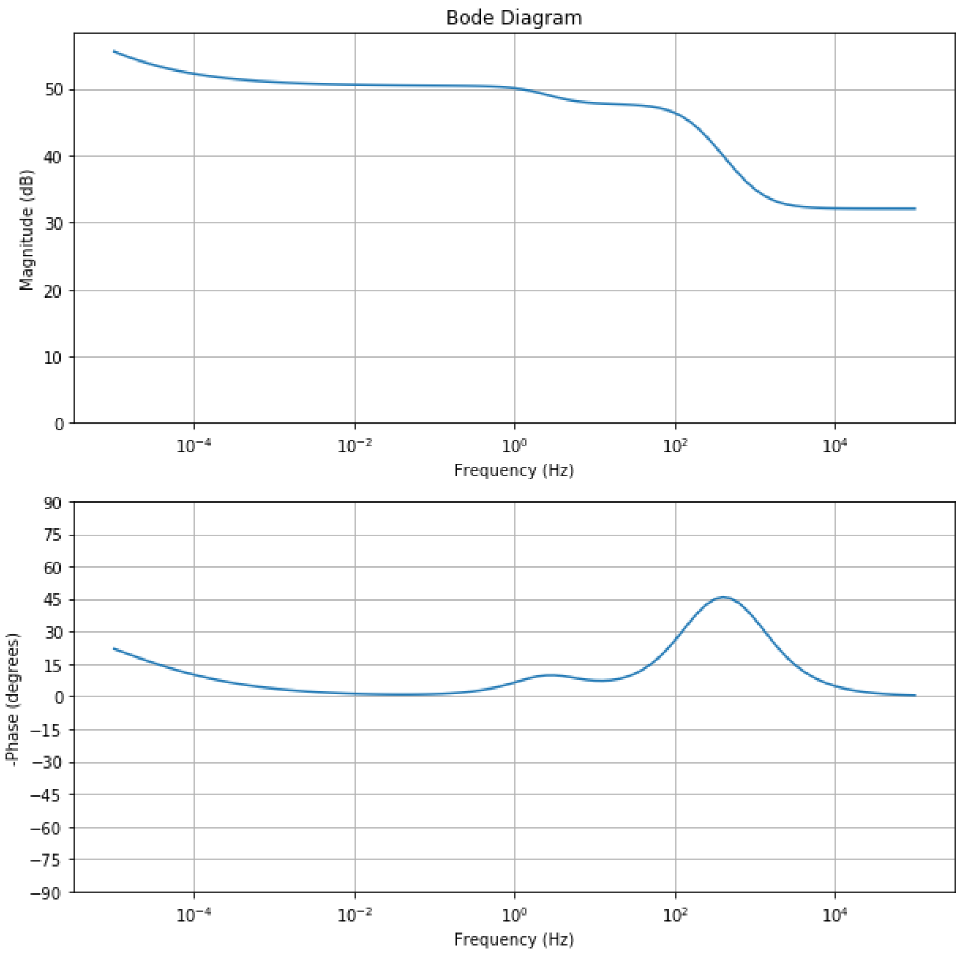
For Constant-Phase Element (CPE)

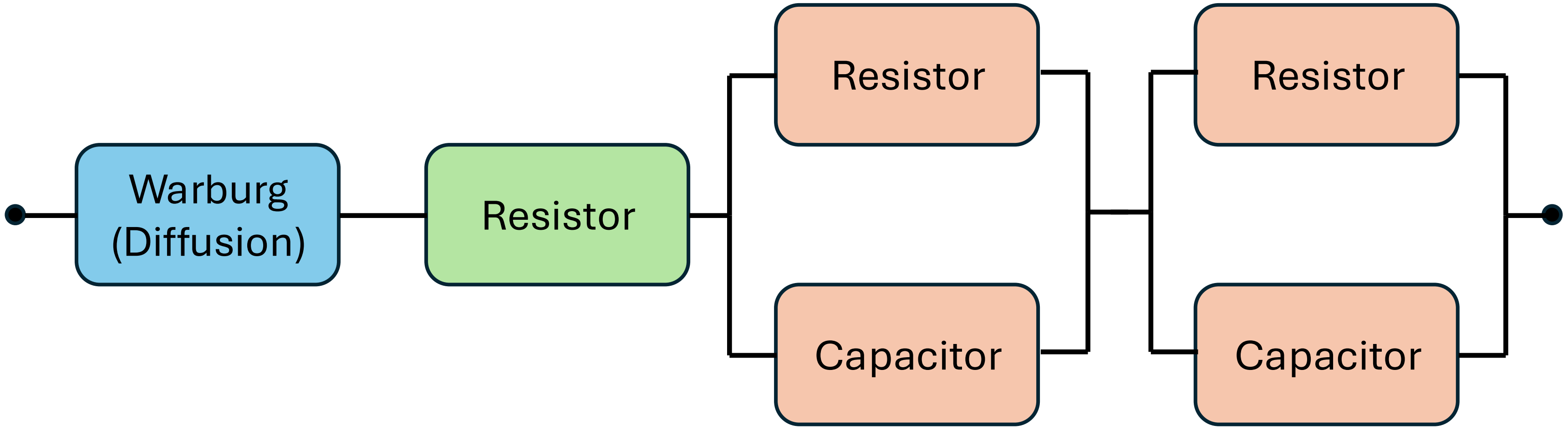
Equivalent Circuit Model

- R is resistance
- C is capacitance
- I is current
- W is Warburg Coefficient
- s = σ+jω
  - σ = 0 is the Fourier Transform
- The Equivalent Circuit Model is the sum of each impedance term

EIS Output


Nyquist Diagram
Direct Current Resistance
Mass Transfer Resistance
-Z imag
Z real
0
100
200
300
400
500


Bode Diagram
Magnitude (dB)
Frequency (Hz)
-Phase (degrees)
10^{-4}
10^{-2}
10^{0}
10^{2}
10^{4}



$$V(t) = 2IW\sqrt{\frac{t}{\pi}}$$

$$V(t) = IR$$

$$V(t) = IR\left(1 - e^{\frac{-t}{RC}}\right)$$

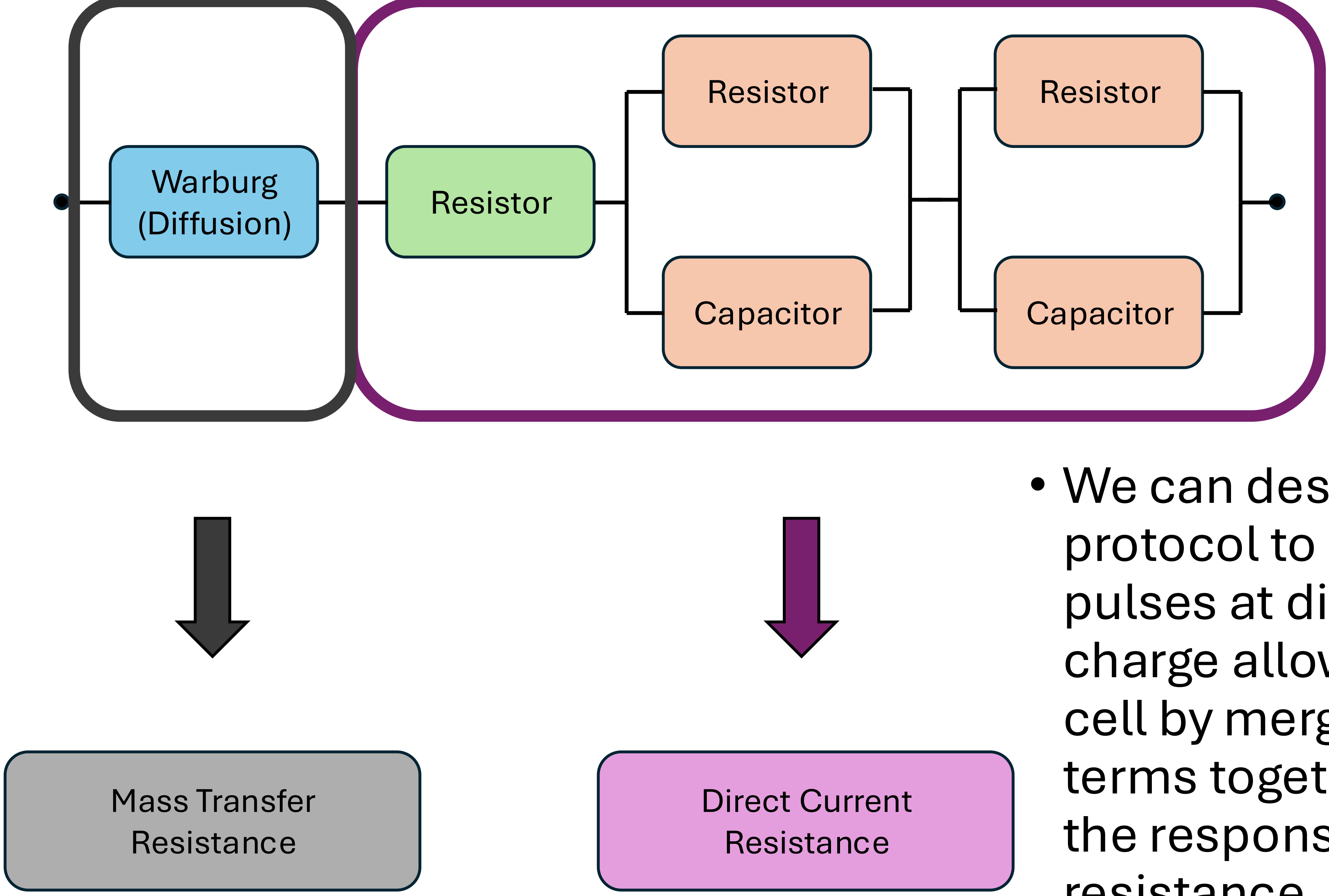
No Analytic solution for Constant-Phase Element (CPE)

- The math behind a step-change in current, when converted back to the time domain

Element	Unit Impulse	Current Step	Voltage Step
Response	1	$I(s) = \frac{I}{s}$	$V(s) = \frac{V}{s}$
Resistor	$Z(s) = R$	$V(t) = IR$	$I(t) = \frac{V}{R}$
Capacitor	$Z(s) = \frac{1}{Cs}$	$V(t) = \frac{It}{C}$	$\delta(t)VC$
Constant Phase Element	$Z(s) = \frac{1}{As^{\alpha}}$	$V(t) = \frac{It^{\alpha}}{A\Gamma(\alpha+1)}$	
Inductor	$Z(s) = Ls$	$V(t) = ILt$	$(t) = \frac{Vt}{L}$
RC	$Z(s) = \frac{R}{RCs+1}$	$V(t) = IR\left(1 - e^{-\frac{t}{RC}}\right)$	$I(t) = \frac{V}{R} + \delta(t)VC$
R-CPE	$Z(s) = \frac{R}{RAs^{\alpha}+1}$	No analytic solution, similar to RC	
Warburg	$Z(s) = \frac{W}{\sqrt{s}}$	$\mathrm{V}(t) = 2IW\sqrt{\frac{t}{\pi}}$	Diverges
For $\alpha \in [0, 1]$, s=σ+iΩ, δ(t) Dirac's delta function			

EIS without the EIS

A poor man’s EIS



- We can design a testing protocol to include current pulses at different states of charge allows for profiling the cell by merging all the DC terms together and measuring the response as mass transfer resistance

Designing a Protocol

- Cell Test Protocols typically charge and discharge a cell up to thousands of times, typically by combining steps that charge or discharge a cell up to thousands of times at a constant current or voltage
- By pausing a cell's discharge and applying a high current pulse, the DC resistance can be measured by the initial change in resistance, and the Mass Transfer resistance by the change in voltage divided by the pulse current
- This can be repeated ~20 times during a cycle to profile the cell as a function of state of charge, and on multiple cycles to see how it evolves as a function of cycle life
- By designing a protocol like this, we can generate the same data for dozens of cells in less time than it would take to achieve this be EIS

What does this buy us?

- By standardizing cell testing to include pulsing cycles, a dataset with tens of thousands of cells and information on specific failure modes can be generated
- One application is to then do a Design of Experiments (DoE) varying some combination of design parameters (anode, cathode, electrolyte, etc.) and learning how they affect the system as a whole
  - The simplest form of a DoE would be an A/B test varying 1 or more variables relative to a control
- Another application is to retroactively go through thousands of past experiments looking at the effects of those same parameters looking for signals to help inform decisions on new experiments

Can we take this further?

- The math behind the method is in the frequency domain using Fourier Transforms, however the data has a time component with state of charge
- By using carefully setting up the math to use state of charge for time, it's possible to then do an Inverse Laplace Transform to see how each element evolves over time
- With this new tool, it is then possible to predict for any current applied what the voltage profile would look like
  - Batteries have upper and lower voltage cutoffs to protect the cells from damage, giving the boundary conditions to solve for capacity

Impedance Model

$$V = IR$$

Add model terms

$$dV = IR + 2IW\sqrt{\frac{t}{\pi}}, t = 3600xC, I = CJ$$

Do lots of algebra

$$\gamma C^3 - \beta^2 C^2 + 2\beta C - \alpha^2 = 0, \gamma = \frac{3600x}{\pi}, \alpha = \frac{dV}{2JW}, \beta = \frac{R}{2W}$$

- For:
  - dV as voltage drop from start of discharge to voltage cutoff in V
  - I as current in A
  - R as resistance in Ω
  - W as Warburg coefficient in Ω/sqrt(s)
  - t as time in s
  - x as fraction discharge
  - J as the current required to move all the electrons in 1 hour
  - C as C-Rate as fraction of J
- What’s left is a cubic expression for C
  - There should only be 1 real root to this equation, though it may be possible to manufacture numbers where this is not true