Data-driven insights into protonic-ceramic fuel cell and electrolysis performance

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Appendix A: Model parameters considered for selection and performance prediction

Below is a detailed explanation of each parameter used in the machine learning models.

1. **Absolute humidity (g/m^3)** - This parameter represents the grams of water vapor per cubic meter of air in the lab at the start of the cell-sintering process. Absolute humidity was derived from the lab relative humidity and temperature, recorded by the HVAC system. An approximation of the Clausius-Clapeyron equation is used to determine the saturation vapor pressure (e_{sat}) in Pascals (Equation S1) [1].

$$e_{\text{sat}} = \frac{\exp(34.494 - \frac{4924.99}{T+237.1})}{(T+105)^{1.57}} \tag{S1}$$

The saturation vapor pressure, derived from the temperature (T) in degrees Celsius, is then used to determine the actual vapor pressure (e) using the relative humidity (RH). The calculation is detailed in Equation S2 [2].

$$e = RH/100 * e_{\text{sat}} \tag{S2}$$

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Finally, the ideal gas law is used to calculate the absolute humidity (AH) in grams of water vapor per cubic meter of air (Equation S3); where MW_{water} is the molecular weight of water (18.02 g/mol), R is the gas constant in J/mol*K, and T is the temperature in Kelvin.

$$AH = \frac{e * MW_{\text{water}}}{R * T} \tag{S3}$$

2. **BCFZY batch** - The batch of BCFZY precursor powder used as the positrode material. Batches are identified by start date of the materials synthesis process.

3. **BCZYYb (wt-%)** - The percentage by weight of BCZYYb precursor powders in the negatrode.

4. **Co-sinter batch** - The group of cells co-sintered together. This categorical data-column is labeled with the co-sintering date to ensure unique identification of each batch.

5. **Co-sinter furnace** - Specifies the furnace used for the high-temperature co-sintering process. The cells were primarily sintered in Furnace 1.

6. **Current at 1.3 V (A/cm^2)** - The current density recorded from the cell at an applied voltage of 1.3 V during electrolysis performance testing.

7. **Days (Co-sinter to test)** - The days elapsed between the co-sintering of the electrolyte and negatrode, and the subsequent performance testing of the cell.

8. **Days (Positrode application to sinter)** - The days elapsed between applying the positrode paste to the cell, and the subsequent sintering of the positrode onto the electrolyte.

9. **Days (Press to spray)** - The number of days elapsed from the pressing of the negatrode pellet to the spraying of the electrolyte onto the pellet.

10. **Days (Spray to sinter)** - The number of days elapsed between the electrolyte spraying onto the negatrode and the half-cell sintering.

11. **Dried before co-sinter** - Whether or not the green negatrode-electrolyte bi-layer was dried in a drying oven at 80–100 °C before the co-sintering process. "Yes" means the cell was dried right before co-sintering. "YesNo" means the cell was dried but not directly before the co-sintering process. "No" means the green body was not placed into a drying oven before the sintering process. Most cells were not dried before co-sintering. Four cells are labeled as "Yes" and three cells are labeled as "YesNo".

12. **Electrolyte application** - Describes the method used to apply the electrolyte onto the negatrode. The electrolyte was applied with a hand sprayer or a Sono-Tek Align ultrasonic spray coater. The majority of electrolytes were applied using the ultrasonic spray coater.

13. **Electrolyte batch** - The batch of electrolyte precursor powders used to form the electrolyte layer. Batches are identified by the start date of the materials synthesis process.

14. **Electrolyte thickness to grain size ratio** - The ratio of the thickness of the electrolyte to the grain size of the electrolyte. Electrolyte thickness was calculated by through measurement under SEM at 10 different spots on three different cross-sectional images of each cell. Grain size was calculated using stereology. Concentric circles were placed over each SEM electrolyte-surface image;the number of intersections between the circles and the grain boundaries was divided by the total length of each circle. This was thrice repeated for each cell.

15. **Electrolyte particle D50 (μm)** - The collective median diameter (D50) of the BCZYYb precursor powders in the electrolyte.

16. **Electrolyte spray batch** - The batch of cells that was sprayed. This categorical data column is labeled as date of spray for the cells to ensure uniqueness.

17. **Electrolyte spray solution** - The spray solution used to suspend the electrolyte precursor powders for spray application onto the negatrode. Of the 88 BCZYYb4411 cells, 80 utilized the standard suspension, which is detailed in the experimental section of our prior work [3]. Six cells utilized a simple suspension which only consisted of polyethylene glycol 400 (PEG), polyvinylpyrrolidone MW 40,000 (PVP), and isopropanol. Two samples used a simple solution with smaller amounts of PVP and isopropanol.

18. **Electrolyte treatment** - Post processing on the electrolyte. Two cells were etched with nitric acid, one cell had the electrolyte lightly sanded using 1200 grit sandpaper, and one cell had the electrolyte polished with micron-sized alumina.

19. **Negatrode functional layer (NFL)** - A parameter representing the specific NFL used in fabricating the cell. Most cells were produced without an NFL. NFL powders consisted of 45 wt-% BCZYYb powders and 55 wt-% Type F (1–2 μm) NiO powder. NFL powders did not contain starch poreformer, as used in the negatrode powders.

20. **Negatrode NiO** - The type of NiO utilized in the negatrode precursor powder. Most negatrode batches consisted of NiO from Alfa Aesar, was over 99% purity, and sieved through a 400 mesh sieve. This powder is listed as 44 μm mesh. Three cells used 1–2 μm Type F NiO from Novamet, indicated as Type F in the data. Two cells were tested with a bi-modal distribution (half 44 μm, half Type F NiO).

21. **Negatrode batch** - The batch of negatrode precursor powders used to form the negatrode of the cell. Negatrode batches consist of the BCZYYb precursors, NiO, and starch (pore former). Batches are identified by the start date of the materials synthesis process. The weight ratios of BCZYYb:NiO:starch are generally around 2:3:1, but there are some slight variations between batches.

22. **Negatrode pellet number** - Indicates the sequence in which the negatrode pellets

were pressed. Binder is combined with negatrode powders, and this mixture is formed into pellets in batches of 9 to 12. Pellet 1 is pressed first and pellet 12 is pressed last. Higher pellet numbers suggest that more water had evaporated from the binder-negatrode mixture before pressing.

23. **Negatrode thickness (mm)** - Thickness of the negatrode in the fabricated cell.

24. **NiO (wt-%)** - The weight % of NiO powder in the negatrode powder mixture. This value had fluctuated by less than one % between batches.

25. **NiO in electrolyte** - A categorical variable indicating if 1 wt-% NiO was added to the BCZYYb precursor electrolyte powders (Yes/No).

26. **NiO particle size (μm)** - Represents the median diameter (D50) of the NiO powder used to fabricate the negatrode.

27. **Peak power density (W/cm^2)** - The peak power density (PPD) of the cell recorded at the beginning of testing. PPD is calculated from a current-voltage curve from OCV to $\sim$0.4 V at a scan rate of 0.001 A/s. This is the target value for the models of fuel cell performance.

28. **Positrode functional layer (PFL)** - Represents the specific PFL used in cell fabrication. Most cells were made without a PFL. In this study, all PFLs were applied using the same spray method as the electrolyte and co-fired with the negatrode and electrolyte.

29. **Positrode paste** - The batch of positrode powder paste, consisting of 2.5 g powder, 0.5 g of 20% Solsperse 28000 in terpineol, and 0.2 g of 5% Heraeus V-006 in terpineol. Occasionally, a half batch with the same ratio of materials was used. Batches are identified by fabrication date and material composition.

30. **Positrode paste age (days)** - The days between when a positrode paste was fabricated and when it was applied to a cell.

31. **Positrode sinter batch** - A categorical variable representing the group of positrodes sintered together in the furnace. Each batch is uniquely identified by its co-sintering date.

32. **Positrode sinter furnace** - The furnace used to sinter the positrode onto the cell.

33. **Positrode sinter temperature (**°C**)** - The temperature used to sinter the positrode, held for five (5) hours. The ramp rate to this temperature was either 1 or 2 °C/min.

34. **Positrode thickness (μm)** - The thickness of the positrode, calculated as the average of 10 measurements from three different SEM images taken after testing. Thickness varied significantly within a single cell.

35. **Silver grid paste** - The silver contact paste used to fabricate the silver grid current collector on each electrode. "Shanghai" refers to DAD-87 paste from Synthetic Resin Research Institute, "Vivtek" to DAD-87 paste from Vivtek, and "FCM" to AG-I, Item # 321201 paste from fuelcellmaterials.com.

36. **Silver spring** - A categorical variable indicating if a bent silver wire was used as a spring to connect the cell to the silver mesh in the test stand (Yes). The spring was cured onto the silver grid, with silver paste applied on top to connect to the electronic load and / or impedance analyzer. "No" indicates a pile of silver paste was used instead.

37. **Sintering neighbor (SN)** - Refers to additional materials placed near the cells in the furnace during high-temperature co-firing of the electrolyte and negatrode. These sacrificial materials were selected to potentially enhance the sintering process.

38. **Sintering temperature (°C)** - The peak temperature reached during the high-temperature co-sintering of the negatrode and electrolyte.

39. **Test air flow (SCCM)** - The amount of air flow to the positrode during performance testing. The first six cells had 50 SCCM of flow, the next five had 100 SCCM, and the remaining cells had 200 SCCM. "Air" for all tests is a synthetic blend of 21% O_2 balanced with Ar flowed through a bubbler at room temperature.

40. **Test stand** - The test stand used for cell performance characterization. Six Mines 4411 cells were tested in Stand 4-2, while the rest were tested in Stand 4-3.

41. **Two-step sinter** - Indicates whether the sintering schedule included a two-step process, where the furnace initially reaches a high temperature, then quickly drops to a slightly lower temperature and for a longer dwell. Options for this parameter are "Yes" or "No." This technique was first outlined by Chen and Wang [4] and applied to PCC devices by Choi et al. [5].

Appendix B: Model parameters tracked but dropped due to 0-variance or collinearities with other parameters.

Below is a detailed explanation of each parameter that was tracked but dropped before the parameter selection process. While these columns were only briefly discussed, they can give researchers ideas of parameters to track for their own cells. Since these columns were dropped, their importance was not studied.

1. **Cell tested** - The name of the cell tested. This column was removed as each cell would represent a unique category.

2. **Date tested** - The date cell testing commenced. This column was removed as each cell would represent a unique category.

3. **Days (Positrode sinter to test)** - The number of days between positrode sintering and cell testing. This parameter is highly collinear with "Days (Co-sinter to test)".

4. **Days (Press to sinter)** - The days between the date of the negatrode pellet pressing and the sintering of the half-cell. The data in this parameter is already captured by the "Days (Press to spray)" and "Days (Spray to sinter)" parameters.

5. **Electrolyte Ce on B-site** - The Ce content on the B-site of the perovskite, expressed as a whole number (e.g., 40, 70) out of 100. Since the models only analyzed BCZYYb4411 cells from Mines, this column had zero variance.

6. **Electrolyte grain size (μm)** - The average electrolyte grain size, calculated using stereology as described in the "Electrolyte thickness to grain size ratio" parmeter. This parameter is captured by the "Electrolyte thickness to grain size ratio" parameter.

7. **Electrolyte material** - The material the electrolyte precursor powders are intended to make. This includes whether or not NiO was added to the electrolyte. Since all cells in the model were BCZYYb4411, this column became redundant with the parameter "NiO in electrolyte".

8. **Electrolyte in negatrode** - The type of electrolyte precursor powder used in the negatrode. All cells in the models used the same electrolyte composition in both the negatrode and electrolyte. Since all model cells were BCZYYb4411, this parameter had zero variance.

9. **Electrolyte spray layers** - The number of electrolyte layers applied to the cell by spraying. Owing to the use of multiple spray techniques, this parameter is somewhat variable and highly collinear with the "Electrolyte thickness" parameter, leading to its removal.

10. **Electrolyte thickness (μm)** - The thickness of the electrolyte of the cell. Thickness was determined by measuring 10 different points on three cross-sectional SEM images of each cell. This parameter was removed as its information is captured by the "Electrolyte thickness to grain size ratio" parameter.

11. **Location** - The location that the cell was tested. For the ML models, all cells were tested at the Colorado School of Mines (Mines), so this column has 0 variance. Figures 7 and 8 include data from three cells tested at Curtin University in Perth, Western Australia, in Prof. Zongping Shao's laboratory (Curtin).

12. **Negatrode binder (%)** - The proportion, in wt-%, of 10% polyvinyl alcohol (PVA) solution mixed with water that is blended into the negatrode powders prior to pellet pressing. This ratio determines the binder content in the mixture. This parameter was removed because it is highly collinear with electrolyte particle size (coincidence).

13. **Negatrode formation** - The fabrication method for the negatrode. As all negatrodes were produced using die pressing, this parameter exhibited zero variance.

14. **Negatrode mass in furnace (g)** - The total mass of negatrode powders in the furnace. This includes all cells and sintering neighbors.

15. **Negatrode-Electrolyte particle D50 (μm)** - Represents the median diameter (D50) of the BCZYYb4411 precursor powders used to fabricate the negatrode. This parameter is highly collinear with NiO particle size, so it was removed from the model.

16. **Normalized green negatrode thickness** - The thickness of the negatrode pellet onto which the electrolyte was sprayed, normalized to the thickness of cells 3 and 5 (7111 cells were excluded from the models). Thickness was calculated based on the weight of negatrode powders in the die. Most cells were fabricated using a 1 1/8" (2.85 cm) die, but for cells using a 2 1/4" (5.7 cm) die, the negatrode pellet weight was divided by four. This parameter is highly collinear with the parameter "Test number" so it was dropped from the model.

17. **Positrode application** - The method used to apply the positrode to the cell. All cells fabricated at Mines used brush painting, resulting in zero variance in this column. The positrodes on cells fabricated at Curtin University were applied using hand spraying.

18. **Positrode material** - The material composition of the positrode. The data in this parameter is already captured by the parameters "BCFZY batch" and "Positrode paste".

19. **Setter** - The setter on which the cells were sintered. The setters comprised of either alumina or a blend of magnesia and yttria-stabilized zirconia (MgYSZ). Setters become contaminated with Ni following repeated use, and turn a bluish color. The contamination levels of the setters were assesed to: New, Lightly used, Used, and Heavily used. It is also noted whether they were ground (sanded down) before sintering.

20. **Shrinkage (%)** - The shrinkage rate of the negatrode-electrolyte bi-layer after co-sintering, expressed as a percentage. Due to missing values, this parameter was removed from the models to increase the number of observations.

21. **Silver grid pattern** - The pattern in which the silver grid was applied to the cells. Most cells used a modified four-line pattern (m4lp), consisting of a cross with two bent lines to minimize the distance between the positrode and silver while reducing the silver area. Four cells had an asterisk pattern, four had a silver dot for connection to the test stand, and seven used a standard four-line pattern. At Curtin, the entire positrode surface was covered with a thin layer of silver mixed with $LaSr_{0.4}Co_{0.6}O_{3-\delta}$ (LSC64) powder. This parameter was dropped from the models because it is highly collinear with the "Test number" parameter.

22. **Spray air flow rate (SLPM)** - The rate at which air flows through the Sono-Tek Impact spray nozzle's air shaping system, measured in standard liters per minute (SLPM). This parameter was set to either 6 or 8 SLPM. This ultrasonic spray parameter, along with four others, was removed from the models due to missing values when hand spraying was used. Dropping these parameters increased the number of observations in the models.

23. **Spray liquid flow rate (mL/min)** - The rate at which the electrolyte solution flows

through the ultrasonic sprayer. For all but one cell, the spray flow rate was set at 0.3 mL/min, while it was adjusted to 0.45 mL/min for the remaining cell. This ultrasonic spray parameter was removed due to missing values.

24. **Spray magnetic stirrer** - Indicates whether a magnetic stirrer was used to mix the electrolyte slurry during the ultrasonic spray process (Yes/No). This ultrasonic spray parameter was removed due to missing values.

25. **Spray Power (W)** - Indicates the power supplied to the Sono-Tek Impact spray nozzle (as part of the ALIGN system), which determines the vibration speed of the nozzle. Higher power settings increase the frequency of these vibrations, enabling the nozzle to generate droplets through high-frequency oscillations ranging from 10,000 to 100,000 Hz. These droplets form on the nozzle surface and are subsequently deposited onto the sample. This ultrasonic spray parameter was removed due to missing values.

26. **Spray size (cm^2)** - Represents the area covered by the spray. A larger area means more cells are sprayed per layer, increasing layer duration. This ultrasonic spray parameter was removed due to missing values.

27. **Test number** - The cell test number. This parameter was removed due to its high collinearity with the "Test air flow (SCCM)" parameter.

28. **Sintering time (hrs)** - The dwell time of the high temperature sintering step, measured in hours.

29. **Starch (wt-%)** - The percentage by weight of the starch measured into the negatrode.

Supplementary Figures

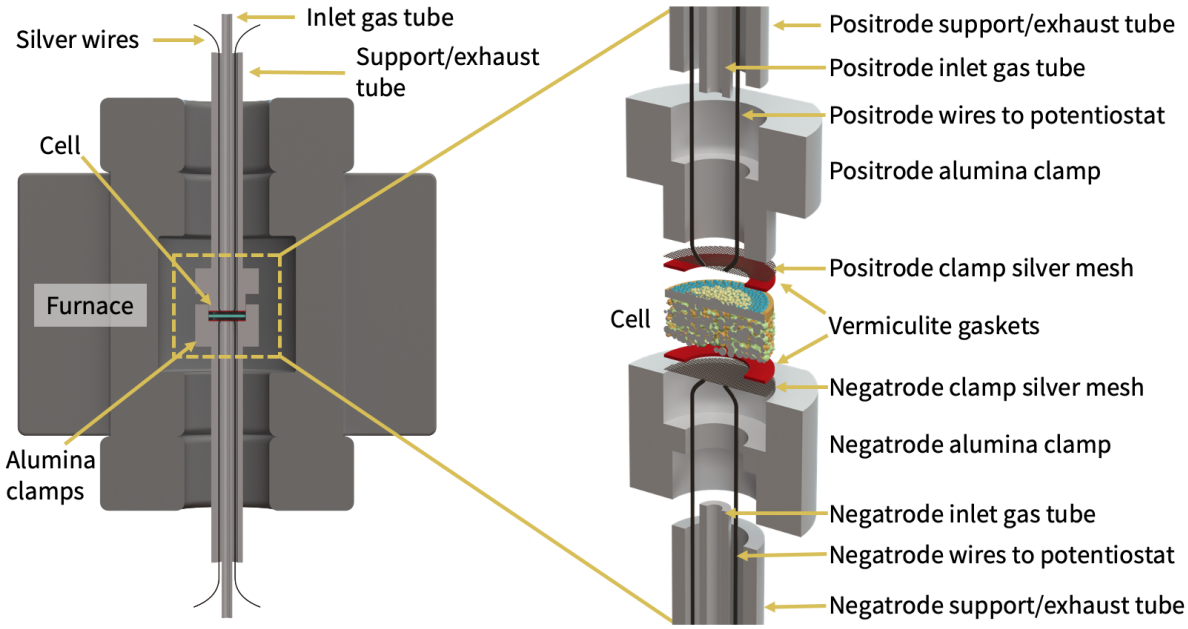

Figure S1: Schematic of the testing setup used at Mines. All tubes and clamps are made from alumina. All current collectors and contact paste are made from silver. Not shown is a hydraulic jack used to apply pressure to form the seal around the cell. Also omitted from the schematic are the silver wires that extend from the cell in a spring-like fashion or silver contact paste to connect the cell with the silver mesh in each clamp. The negatrode and positrode silver wires electronically connect the clamp meshes with the potentiostat leads. The underlying schematic is courtesy of Tyler Burt.

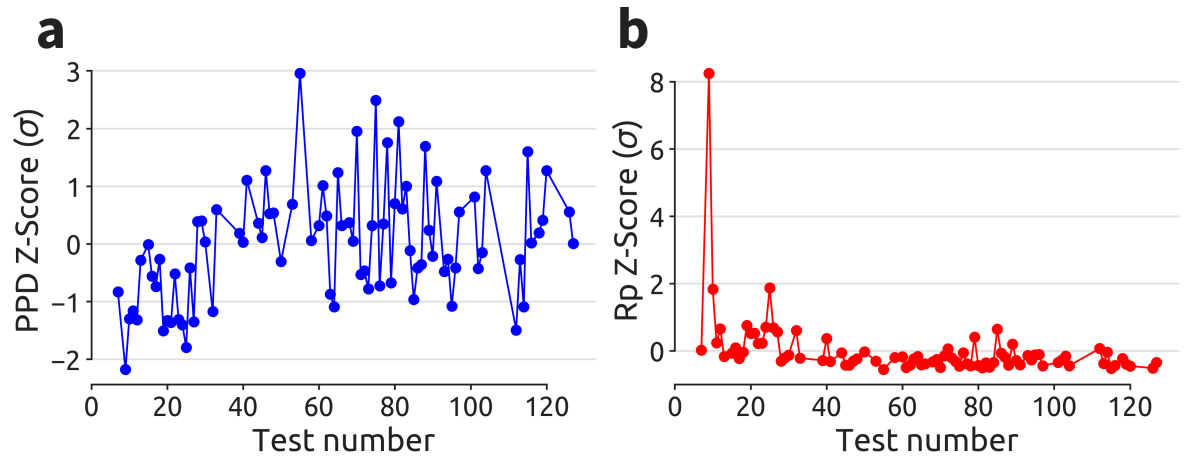

Figure S2: Standard deviation (σ) of a) peak power density (PPD) and b) polarization resistance (Rp) for cells in the fuel cell performance GP model. No PPD data points exceeded the 3σ threshold for dropping a data point. Cell nine exhibited an Rp exceeding 3σ and was excluded, resulting in a final analysis of 86 cells.

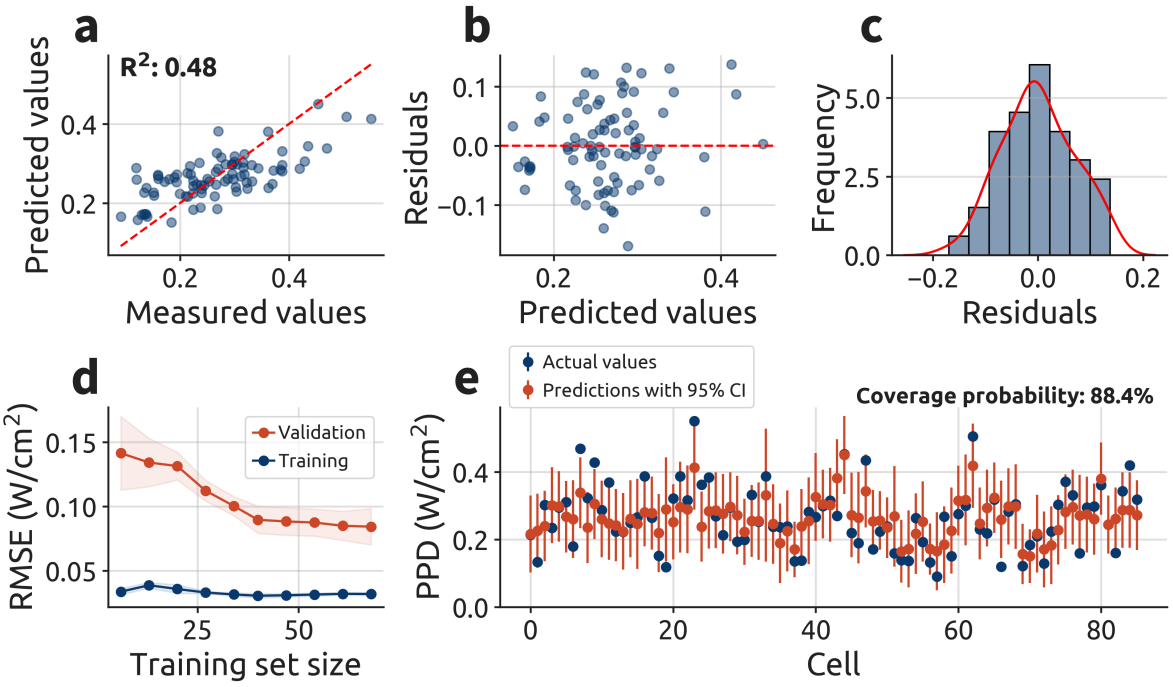

Figure S3: Goodness-of-fit evaluators for the fuel cell performance GP model after 5-fold cross-validation. a) Predicted vs. measured peak power density (PPD) values (W/cm^2). The linear fit shows an R^2 of 0.48, indicating a moderate fit of the model to unseen data. b) Residuals vs. model predicted values. c) Residual distribution, approximately Gaussian around zero, indicating no model bias. d) Learning curves showing a decreasing validation curve with a larger test size, which indicates robust model learning. The gap between validation and training curves suggests the model is overfitting the data. e) Coverage plot comparing predicted PPD (with 95% confidence intervals) to measured values. The model has a coverage probability of 88.4%, meaning 88.4% of all the predictions had the measured value within the predicted 95% confidence interval. Large error bars reflect the difficulty in precisely predicting cell performance.

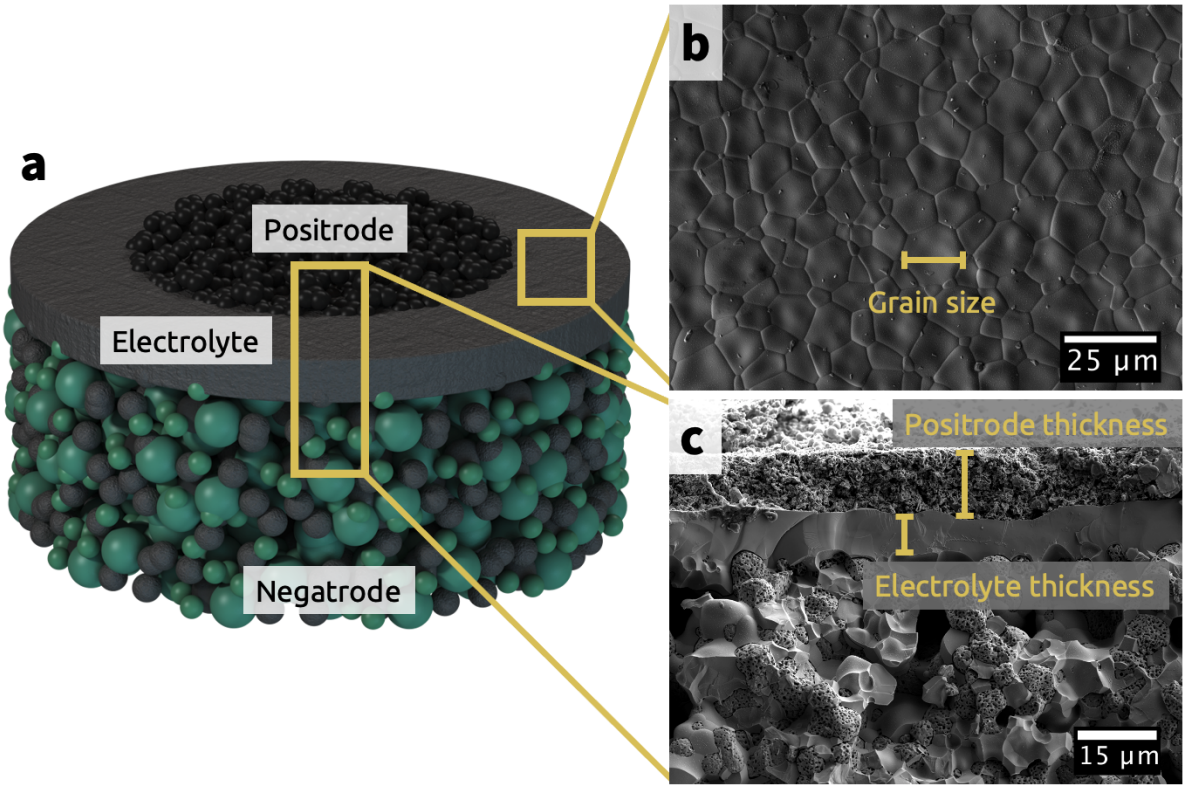

Figure S4: Visualization of key cell parameters. a) Schematic of a button cell. b) Top-view SEM image of the electrolyte, showing grain size. Grain size was calculated via stereology as described in Section 2.4. c) Cross-sectional SEM image of the positrode, electrolyte and negatrode indicating the electrolyte and positrode thickness. Thickness measurements were performed using the methodology detailed in Section 2.4.

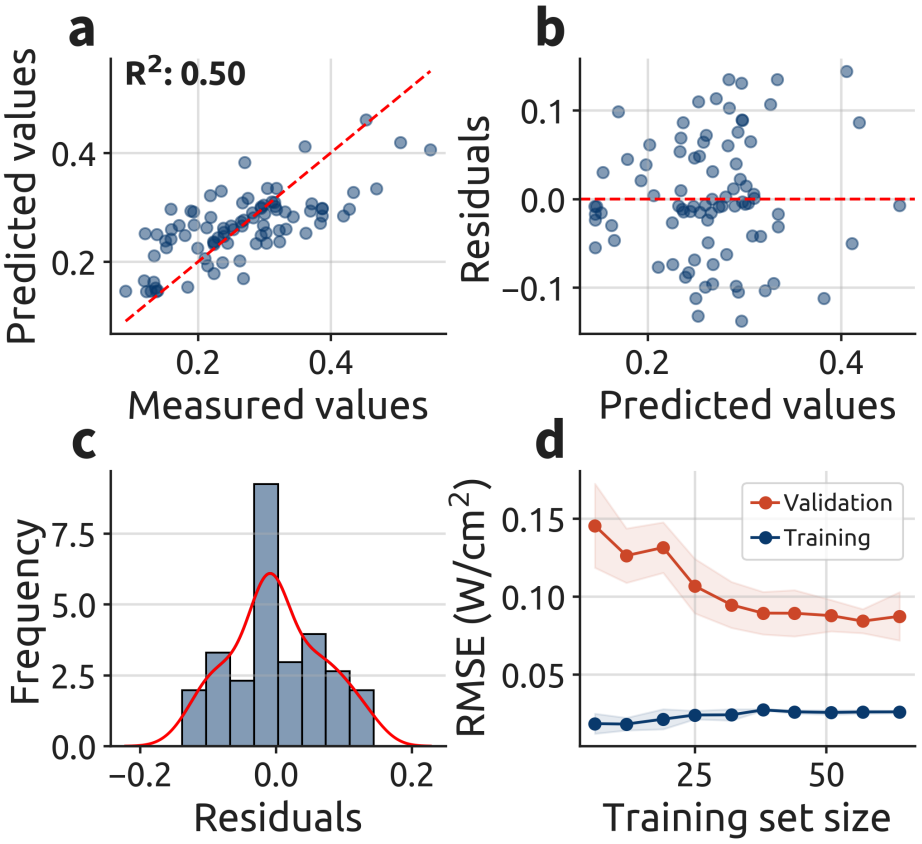

Figure S5: Goodness-of-fit metrics for the fuel cell RFR model after five-fold cross-validation. a) Predicted vs. measured PPD values (W/cm^2), with a linear fit showing an R^2 of 0.50, indicating a moderate fit. b) Residuals vs. model predicted values. c) Residual distribution, approximately Gaussian around zero, indicating no model bias. d) Learning curves, with a decreasing validation curve as test size increases and a larger gap between validation and training curves, suggesting overfitting. The gap is larger than that of the GP model, indicating the RFR model may have more overfitting. The training curve converges toward the validation curve, as expected with more data points.

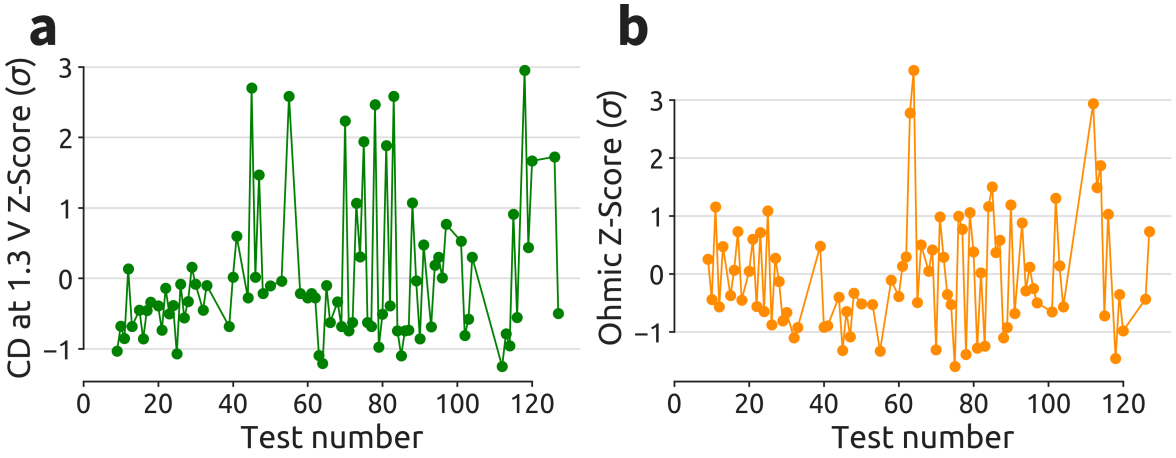

Figure S6: Standard deviation (σ) of a) current density (CD) at 1.3 V and b) ohmic resistance for cells in the electrolysis GP model. No CD data points exceeded the 3σ threshold for dropping a data point. Cell 64 exhibited an ohmic resistance exceeding 3σ and was excluded, resulting in a final analysis of 84 cells.

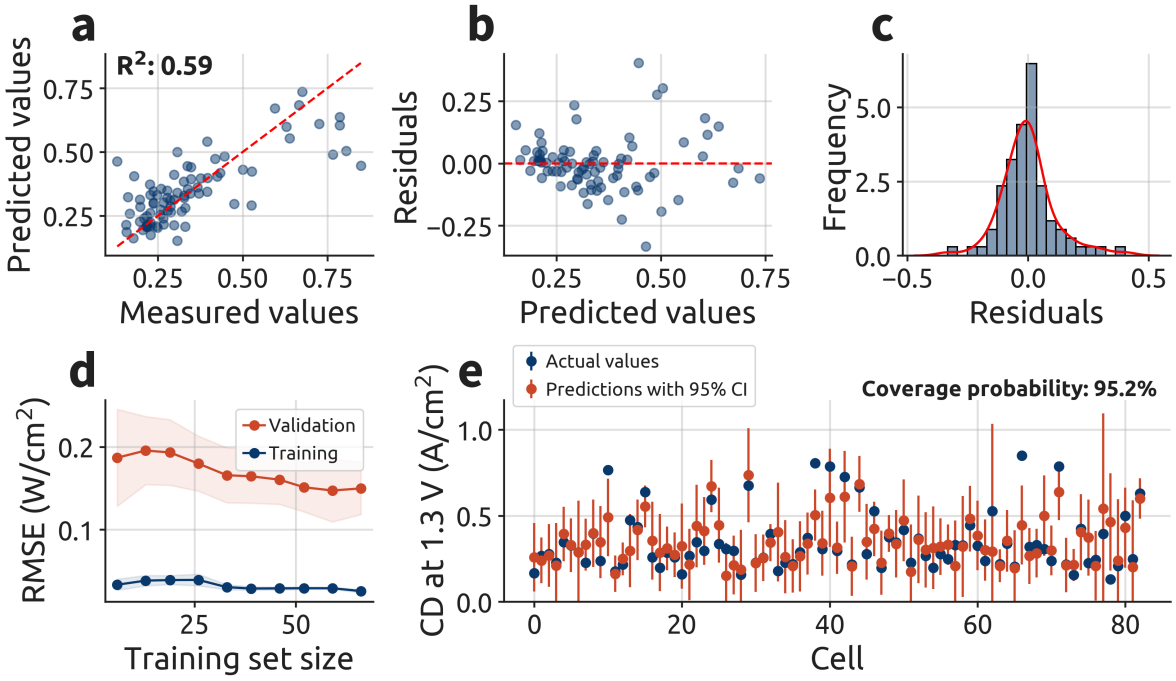

Figure S7: Goodness-of-fit evaluators for the electrolysis performance GP model after 5-fold cross-validation. a) Predicted vs. measured current density (CD) at 1.3 V (A/cm^2). The linear fit shows an R^2 of 0.59, indicating a moderate to good predictive performance.b) Residuals vs. model predicted values. c) Residual distribution, approximately Gaussian around zero, indicating no model bias. d) Learning curves, showing a slight decrease in the validation curve with increasing test size, and a large gap between validation and training curves, suggesting overfitting. Overfitting is expected from models with small sample sizes e) Coverage plot comparing predicted CD at 1.3 V (with 95% confidence intervals) to measured values, with 95.2% coverage. Large error bars indicate the difficulty of precisely predicting cell performance.

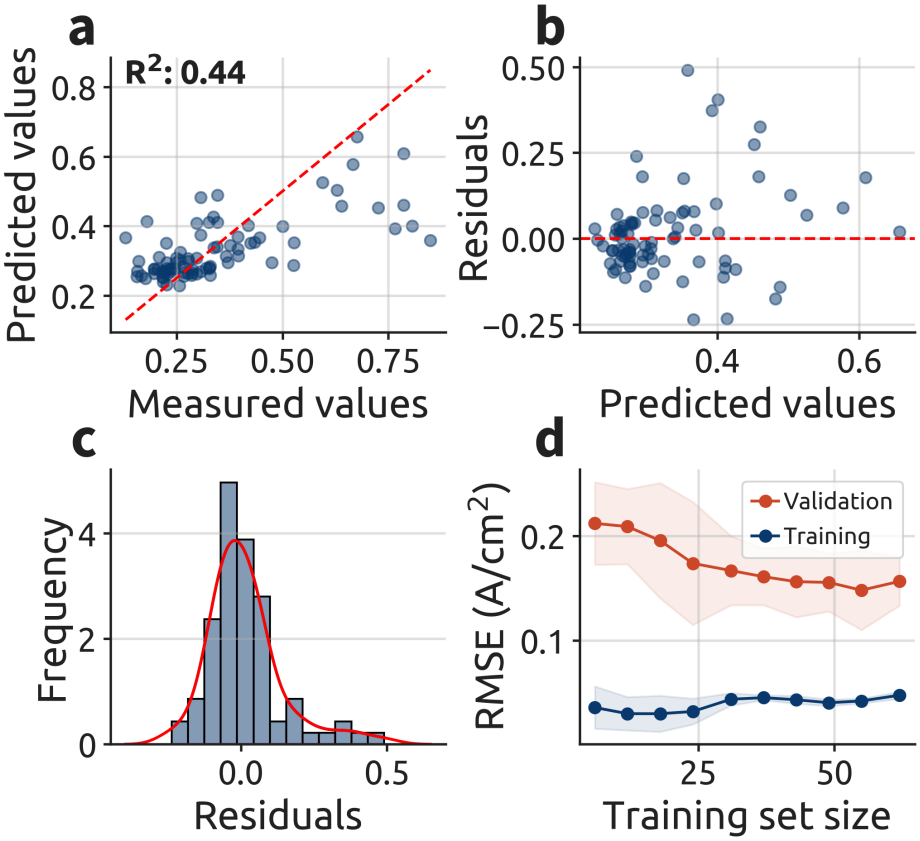

Figure S8: Goodness-of-fit metrics for the electrolysis RFR model after 5-fold cross-validation. a) Predicted vs. measured CD at 1.3 V(A/cm^2). The linear fit shows an R^2 of 0.48, indicating a moderate fit of the model to unseen data. b) Residuals vs. model predicted values. c) Residual distribution, with a slight tail at higher residuals indicating trouble fitting higher performance cells. d) Learning curves, showing a slight decrease in the validation curve with increasing test size, and a large gap between validation and training curves, suggesting overfitting. The gap is smaller than that of the GP model, indicating the RFR model may have less overfitting. The training curve converges toward the validation curve, as expected with more data points.

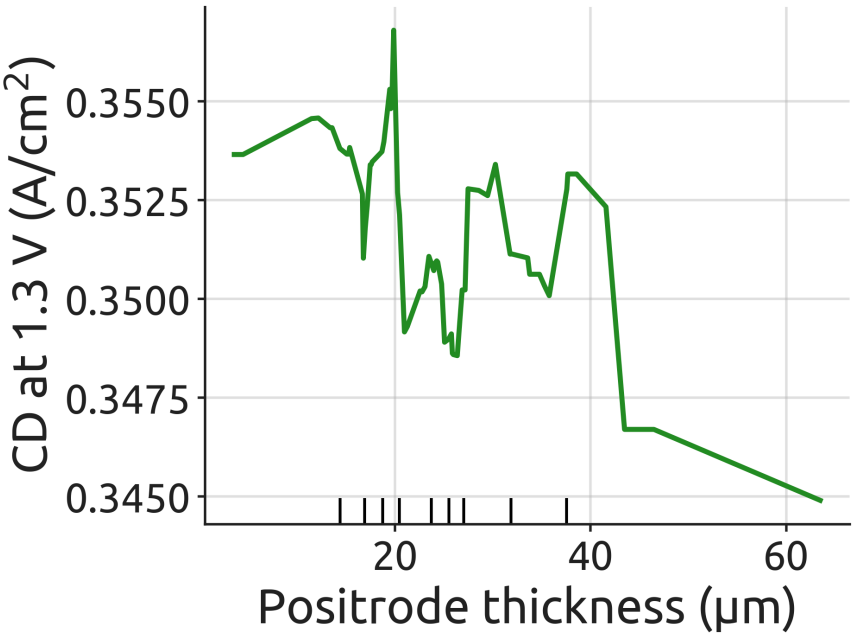

Figure S9: PDP for the "Positrode thickness (μm)" parameter calculated by the electrolysis performance RFR model.

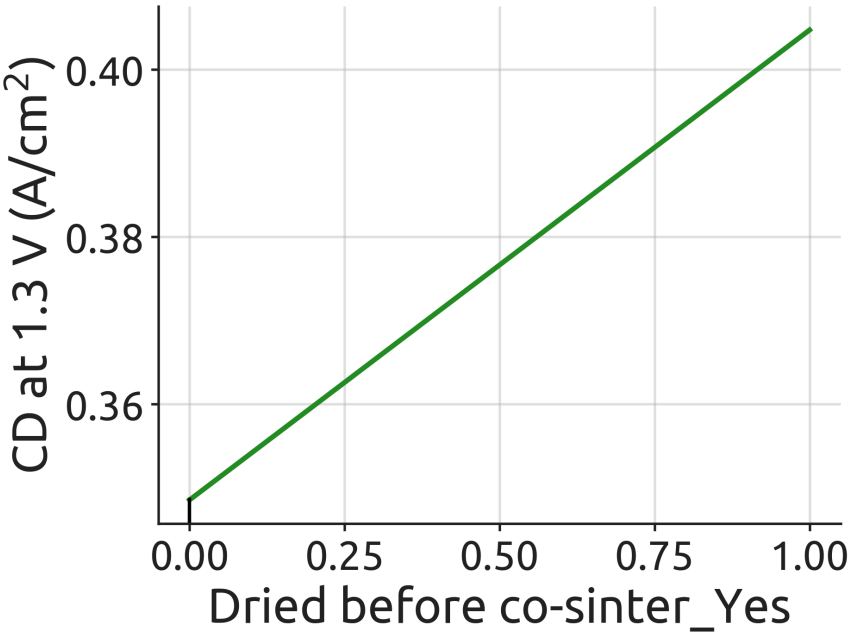

Figure S10: PDP for the encoded parameter "Dried before Co-sinter_Yes" calculated by the RFR model on electrolysis performance. The PDP is linear because there are two options 0 (False) and 1 (True).

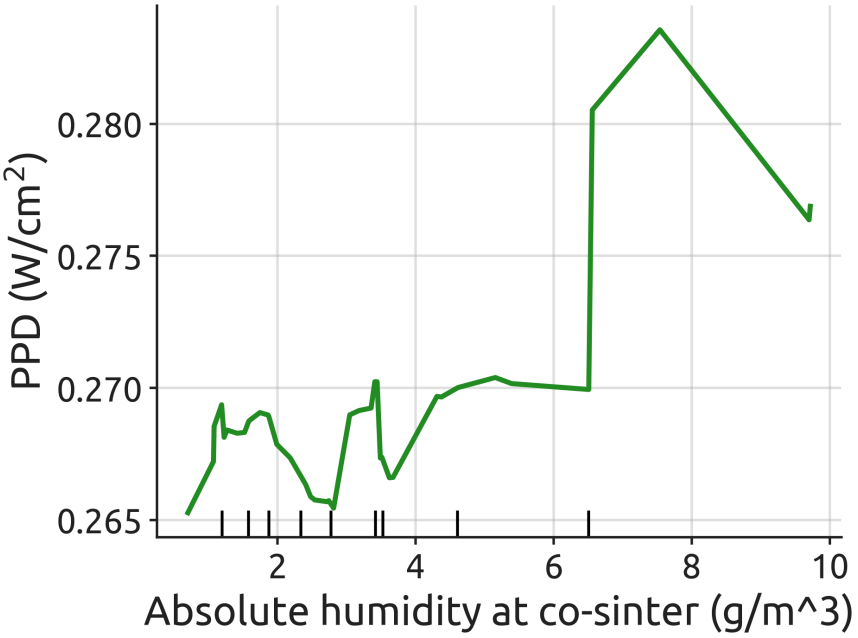

Figure S11: PDP for the "Positrode thickness (μm)" parameter calculated by the electrolysis performance RFR model.

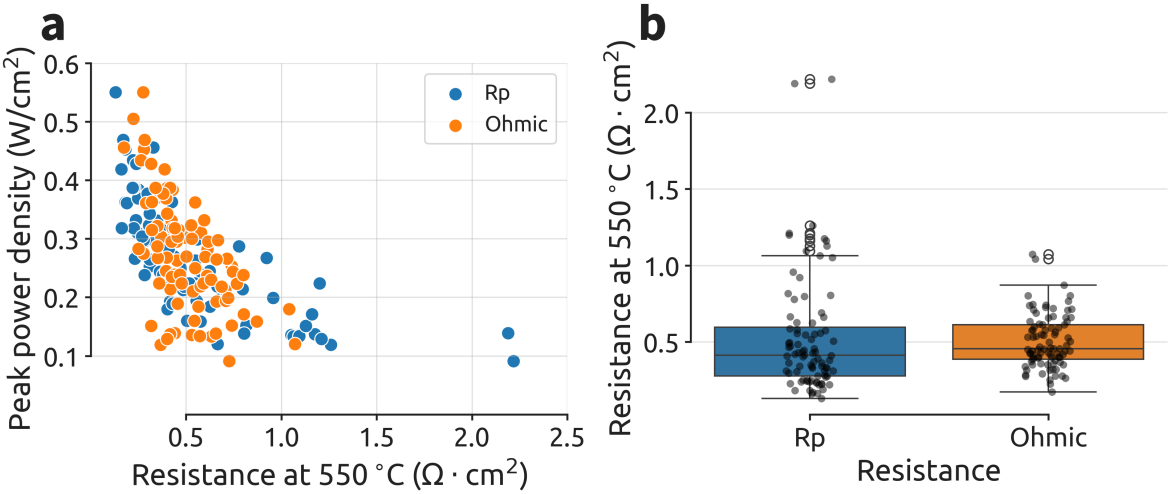

Figure S12: Ohmic and polarization resistance for the 88 cells tested at Mines and Curtin. a) Scatter plot of the resistances. b) Box-and-whisker plots of the resistances. All data were obtained at 550 °C.

Supplementary Tables

Table S1: Properties of the precursors used in the preparation of SSRS BCZYYb4411 electrolyte or negatrode powder.

Compound	**Supplier**	**Purity (%)**	**Particle Size (μm)**
$BaCO_3$	Alfa Aesar	99.8	1
CeO_2	Alfa Aesar	99.9	5
ZrO_2	Sigma Aldrich	99	5
Y_2O_3	Alfa Aesar	99.999	<10
Yb_2O_3	Alfa Aesar	99	4
Coarse NiO	Alfa Aesar	99	<44
Type F NiO	Novamet	99	1-2
Isopropanol	Pharmaco	99	-

Table S2: Properties of the precursor nitrates and polymers for BCFZY sol-gel processing

Compound	**Supplier**	**Purity (%)**
$Ba(NO_3)_2$	Alfa Aesar	99
$Co(NO_3)_2 \cdot 6H_2O$	Alfa Aesar	98–102
$Fe(NO_3)_3 \cdot 9H_2O$	Alfa Aesar	98
Zirconyl nitrate solution	Sigma Adlrich	99
$Y(NO_3)_3 \cdot 6H_2O$	Alfa Aesar	99.9
Ethylenediaminetetraacetic acid (EDTA)	Alfa Aesar	99.4
Citric acid	Alfa Aesar	99–102
Ammonium hydroxide	Pharmaco	28–30

Table S3: Properties of the polymers used to fabricate negatrode pellets and the electrolyte spray solution

Compound	**Supplier**	**Purity (%)**	**Molecular weight (MW)**
Polyvinyl alcohol (PVA)	Alfa Aesar	-	88,000 - 97,000
V-006	Heraeus	-	-
alpha terpineol	Alfa Aesar	96	-
Polyethylene glycol (PEG)	Alfa Aesar	-	400
polyvinylpyrrolidone (PVP)	Alfa Aesar	-	40,000

Table S4: Parameters dropped from the model before selection. Each column lists the reason for exclusion. "Missing values" indicates data was missing, requiring the removal of cells from the model. "Unique" means the parameter consisted of only unique categorical values. "Zero variance" indicates the parameter did not vary in the final model. "Complete collinearity" means all information from one parameter was captured by another. "High collinearity" refers to pairs of parameters with Pearson coefficients above 0.5.

Missing values	Unique	Zero variance	Complete collinearity	High collinearity
Spray air flow rate (SLPM)	Cell tested	Location	Days (Press to sinter) [Days (Press to spray) & Days (Press to sinter)]	Negatrode-Electrolyte particle D50 (μm) [NiO particle size (mum)]
Spray liquid flow rate (mL/min)	Date tested	Positrode application	Sintering time (hrs) [Two-step sinter]	Normalized green negatrode thickness [Test number]
Spray magnetic stirrer		Electrolyte Ce on B-site	Electrolyte material [NiO in electrolyte]	Negatrode mass in furnace (g) [Test number]
Spray Power (W)		Negatrode formation	Electrolyte grain size (μm) [Electrolyte thickness to grain size ratio]	Starch (wt-%) [BCZYYb (wt-%)]
Spray size (cm2)		Electrolyte in negatrode	Electrolyte spray layers [Electrolyte thickness (μm)]	Electrolyte thickness (μm) [Electrolyte thickness to grain size ratio]
Shrinkage (%)			Positrode material [Positrode paste & BCFZY batch]	Negatrode binder (%) [Electrolyte particle D50 (μm)]
Setter				Days (Positrode sinter to test) [Days (Co-sinter to test)]
				Silver grid pattern [Test number]
				Test number [Test air flow (SCCM)]

Table S5: Results of parameter selection for the fuel cell GP model. Each column is titled based on the effect of removing that parameter from the model.

Model got worse	Slightly worse	No change	Slightly better	Model improved
Positrode paste	Positrode thickness (μm)	NiO (wt%)	Positrode sinter temperature (°C)	Absolute humidity at co-sinter (g/m^3)
Co-sinter batch	Negatrode batch	Silver grid paste	Sintering temperature (°C)	Positrode paste age (Days)
NiO particle size (μm)	Electrolyte batch	PFL	NFL	
Electrolyte thickness to grain size ratio	BCFZY batch	Electrolyte spray batch	BCZYYb (wt%)	
Positrode sinter batch	Two-step sinter	Dried before co-sinter	NiO in electrolyte	
Days (Spray to sinter)	Positrode sinter furnace	Sintering neighbor	Negatrode thickness (mm)	
Days (Positrode application to sinter)	Electrolyte treatment	Negatrode NiO	Days (Co-sinter to test)	
		Negatrode pellet number	Test air flow (SCCM)	
		Electrolyte particle size D50 (μm)	Silver spring	
Legend:		Electrolyte application		
Electrolyte		Co-sinter furnace		
Negatrode		Electrolyte spray solution		
Positrode		Days (Press to spray)		
Co-sintering		Test stand		
Testing				

Table S6: Fabrication parameters and peak power density (PPD) for the top 25 performing cells. Higher-performing cells tend to have longer spray-to-sinter intervals, with 10 out of 14 cells sintered more than 10 days after spraying represented in this table. These cells primarily come from a few electrolyte spray and co-sinter batches.

Test number	Electrolyte spray batch	Co-sinter batch	Days (Spray to sinter)	PPD (W/cm2)
55	24Jan22	28Feb22	35	0.550
75	24Jan22	9Mar22	44	0.505
81	24Jan22	17Mar22	52	0.469
70	24Jan22	28Feb22	35	0.453
78	8Mar22	27Mar22	19	0.434
88	10Jun22	16Jun22	6	0.428
115	22Sep23	2Oct23	10	0.419
46	20Oct21	22Oct21	2	0.387
104	29Jun22	22Nov22	146	0.387
120	27Dec23	27Dec23	0	0.387
65	6Apr22	10Apr22	4	0.384
41	20Oct21	22Oct21	2	0.371
91	3Aug22	21Aug22	18	0.369
61	11Apr22	13Apr22	2	0.362
83	24Jan22	17Mar22	52	0.361
101	10Jun22	16Jun22	6	0.343
80	21Apr22	23Apr22	2	0.332
53	24Jan22	26Jan22	2	0.331
82	29Jun22	3Jul22	4	0.323
33	18Nov20	18Nov20	0	0.322
126	27Dec23	27Dec23	0	0.318
97	8Mar22	23Mar22	15	0.318
48	20Oct21	22Oct21	2	0.316
47	20Oct21	25Oct21	5	0.315
62	11Apr22	13Apr22	2	0.311

Table S7: Summary of model error values. The units for root mean squared error (RMSE) and mean absolute error (MAE) are W/cm^2 for fuel cell models and A/cm^2 for electrolysis models. R^2 and out-of-bag (OOB) error (derived from R^2) range from $-\infty$ to 1, with 1 indicating a perfect fit. Lower, negative log-likelihood (NLL) values (higher absolute value) indicate a better fit.

Model	RMSE	MAE	R^2	NLL	OOB
Fuel cell GP	0.068	0.055	0.48	-1.250	N/A
Fuel cell RFR	0.067	0.053	0.50	N/A	0.42
Electrolysis GP	0.109	0.075	0.59	-0.888	N/A
Electrolysis RFR	0.127	0.088	0.44	N/A	0.45

Table S8: Summary of Hyperparameters used in the Random Forest Regressor models. For more details on each parameter, see Section 2.6.

Model	n_estimators	min_samples_split	min_samples_leaf	max_features
Fuel cell	99	2	1	0.205
Electrolysis	72	2	1	0.103

Table S9: Results of parameter selection on the electrolysis GP model. Each column is titled for the effect of removing the column from the model. The table column "Mixed" signifies that some performance metrics improved while others got worse upon the removal of the parameter.

Model got worse	**Slightly worse**	**Mixed**	**No change**	**Slightly better**	**Model improved**
Electrolyte spray batch	Sintering neighbor	Positrode sinter batch	Electrolyte spray solution	Positrode sinter furnace	Negatrode pellet number
Negatrode batch	Co-sinter batch	Dried before co-sinter	BCFZY batch	Electrolyte particle size D50 (μm)	BCZYYb (wt%)
Positrode thickness (μm)	NiO particle size (μm)	NiO (wt%)	Electrolyte application	Silver spring	Silver grid paste
Electrolyte thickness to grain size ratio	Co-sinter furnace	PFL	NiO in electrolyte	Electrolyte batch	Positrode paste age (Days)
Two-step sinter	Electrolyte treatment	Negatrode thickness (mm)		Negatrode NiO	Positrode sinter temperature (°C)
Absolute humidity at co-sinter (g/m^3)				NFL	Positrode paste
Days (Spray to sinter)		Legend: Electrolyte		Test air flow (SCCM)	Sintering temperature (°C)
Positrode application to sinter		Negatrode			Days (Press to spray)
Days (Co-sinter to test)		Positrode			
Test stand		Co-sintering			
		Testing			

Table S10: Exponential and reciprocal fit results for fuel cell and electrolysis performance as a function of ohmic and polarization resistance. Bold font indicates the better fit. Since the exponential and reciprocal models have different complexities (number of variables), the Akaike information criterion (AIC) and Bayesian information criterion (BIC) were used to select the optimal fit. AIC and BIC penalize more complex models, favoring simpler ones [6]. Variables a and b are the fitting parameters from Equation 1. Variable c is the fitting parameter for Equation 2.

Exponential	**a) PPD vs. Ohmic**	**b) PPD vs. Rp**	c) CD vs. ohmic	**d) CD vs. Rp**
R^2	**0.38**	**0.63**	0.72	**0.24**
AIC	**-451**	**-497**	-406	**-318**
BIC	**-447**	**-492**	-401	**-313**
a	**0.53**	**0.46**	1.40	**0.59**
b	**1.38**	**1.15**	2.98	**1.12**
Reciprocal	a) PPD vs. Ohmic	b) PPD vs. Rp	**c) CD vs. ohmic**	d) CD vs. Rp
R^2	0.25	0.29	**0.74**	0.20
AIC	-437	-442	**-413**	-315
BIC	-434	-439	**-410**	-313
c	8.63	11.15	**6.17**	8.55

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