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A physics-based data-driven model for electrolyte supported SOCs

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Motivation

- Solid oxide cells operating at high-temperature (*e.g.*, 600-900°C) display high efficiencies, and convert chemicals into electricity in fuel cell mode. While the reverse process occurs in electrolyser mode.
- Critical challenges are their long-term durability and material degradation processes, and thermal management.
- Speeding up stack performance simulations by coupling multi-scale modelling approaches [1,2] and machine learning algorithms can facilitate to study long-term operating effects.

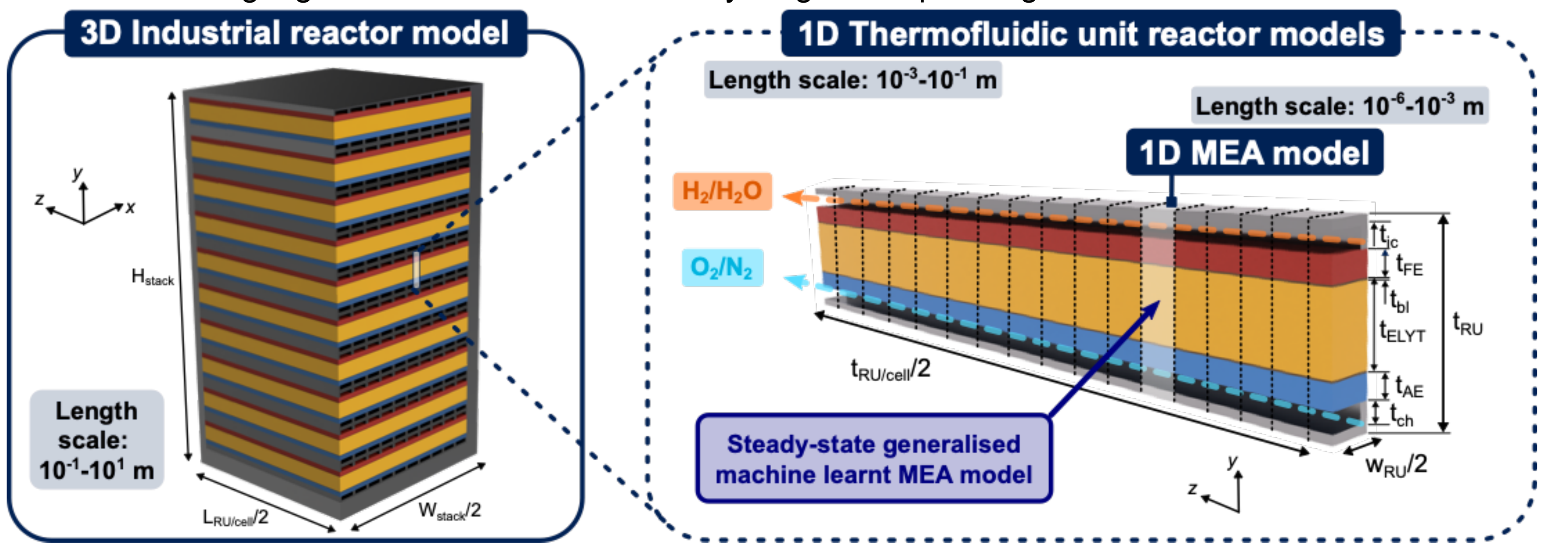


Figure 1. Hierarchical multiscale modelling approach [1,2] and domains: a quarter of a stack and half of a repeating unit (RU).

Towards lifetime modelling

- Leveraging machine learning (ML) algorithms to develop an industrial scale simulator of faster evaluation by incorporating an MEA machine learnt model in a hierarchical multiscale framework.
- Feature engineering analysis of Multiphysics problem to obtain a reduced number of inputs while maintaining physical interpretation: physics-based machine learnt model
- Random forests (RF) are popular ML methods due to their proven accuracy, stability, and ease of use, and provide straightforward methods for feature selection.
- RF regressors (RFR): non-parametric ensemble regression → decision trees as base classifiers, using
- Bootstrapping: random selection of data subsets → overcome over-fitting and reduce prediction error.

Modelling methodology

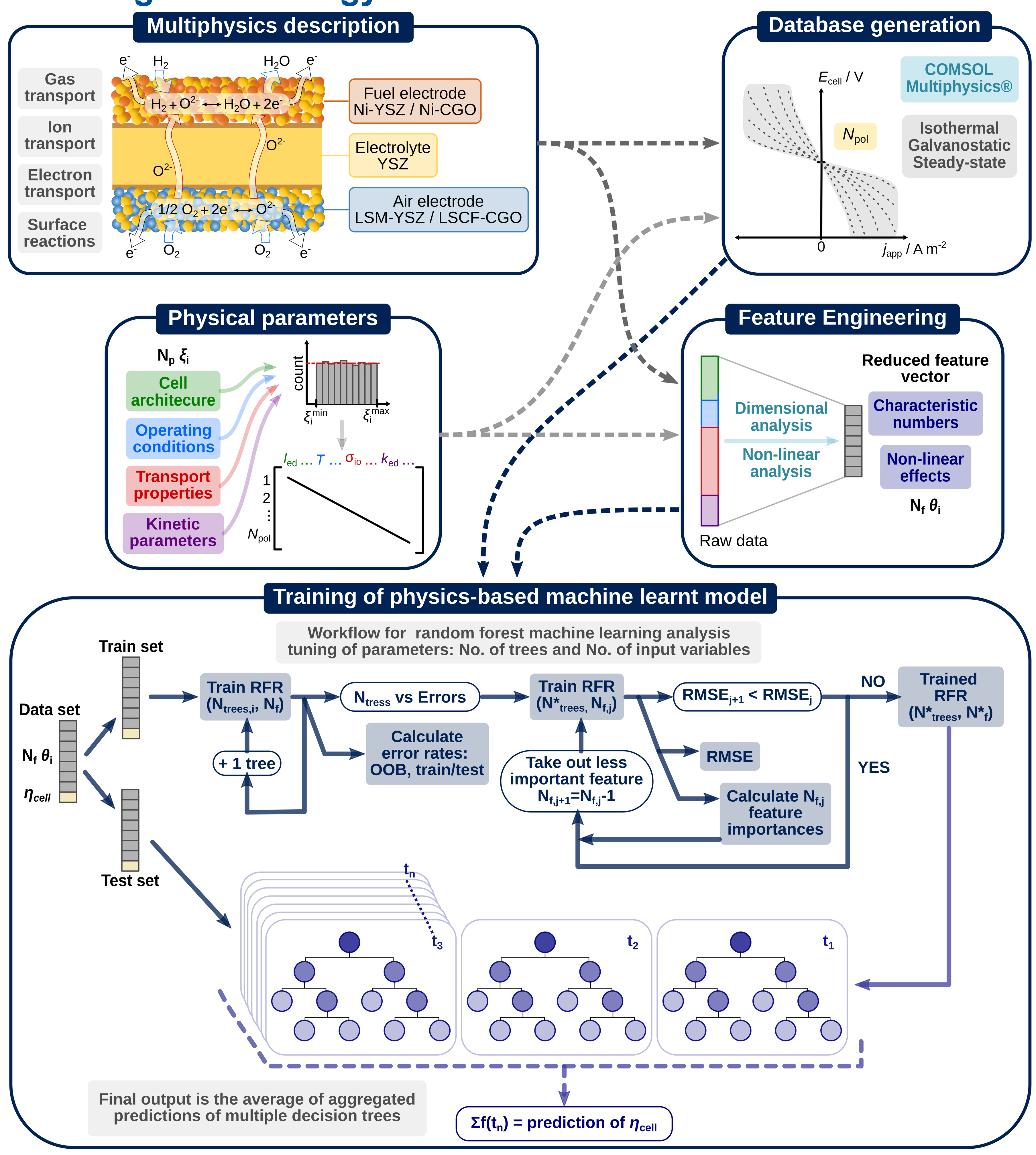


Figure 2. Methodology of physics-based machine learnt model:

i. Generate the mathematical description of 1D steady-state SOC Multiphysics problem (PDEs) and implement in COMSOL Multiphysics®.
ii. A total (N_p) of physical parameters (ξ_i) with upper and lower bounds are used to generate random combinations and solve the problem in COMSOL, obtaining the cell overpotential (η_{cell}, output target) for a total of polarisation cases (N_{pol}).
iii. Using feature engineering (*i.e.*, dimensional and nonlinear effects analyses) to analyse the mathematical description and produce a reduced vector of features (θ_i) containing characteristic numbers and non-linearities.
iv. All ξ_i values are used to evaluate the corresponding θ_i values for N_{pol} cases.
v. Splitting the data (*i.e.*, θ_i and η_{cell}) into train and test data sets.
vi. Train the RFR with all features for an increasing number of tress (N_{trees}) and compare Out-of-Bag (OOB), train and test error rates.
vii. Take out features by considering their permutation importances and RMSE of RFRs trained with N_f number of features.
viii. The final RFR is trained with N^*_{tress} and N^*_f and used to predict the cell overpotential for the test data set.

Training/testing physics-based machine learnt model

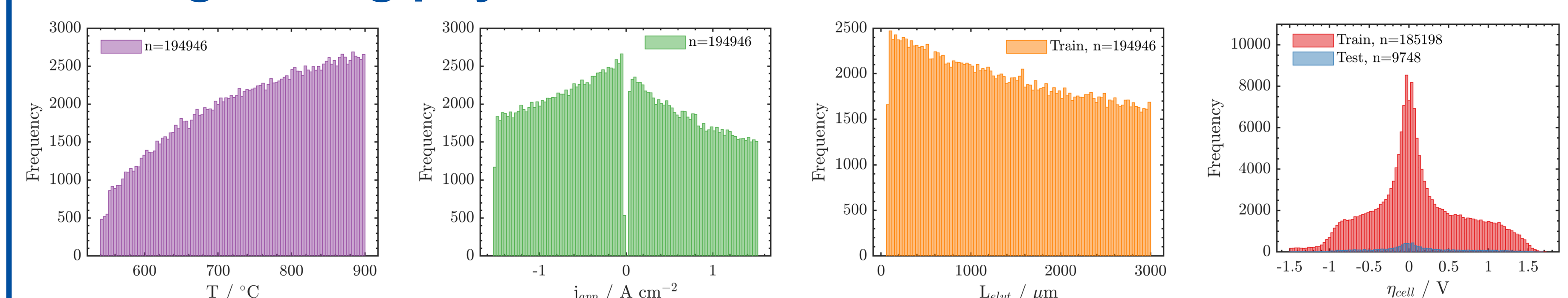
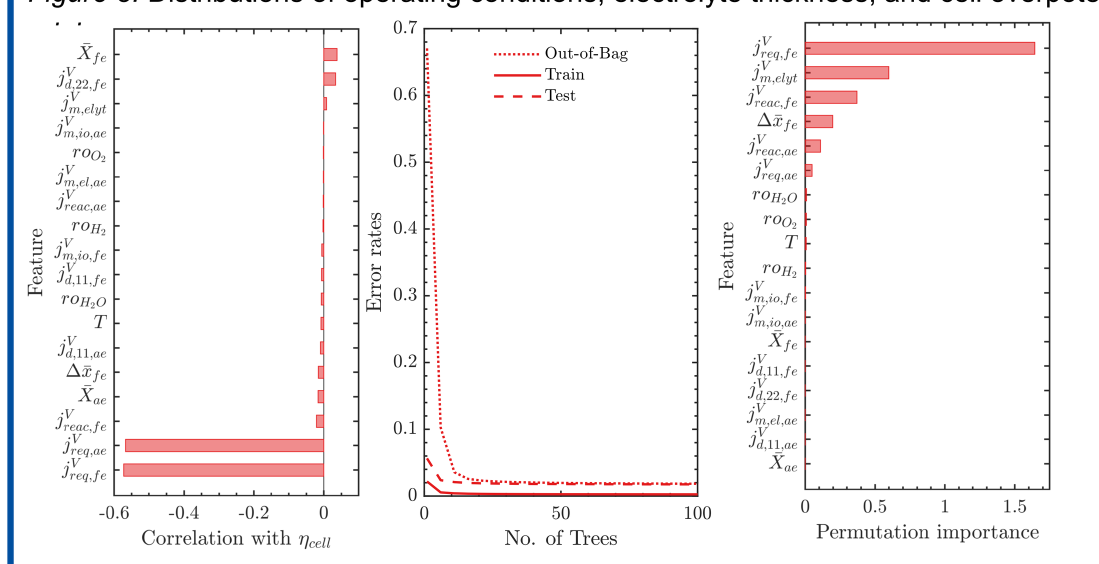
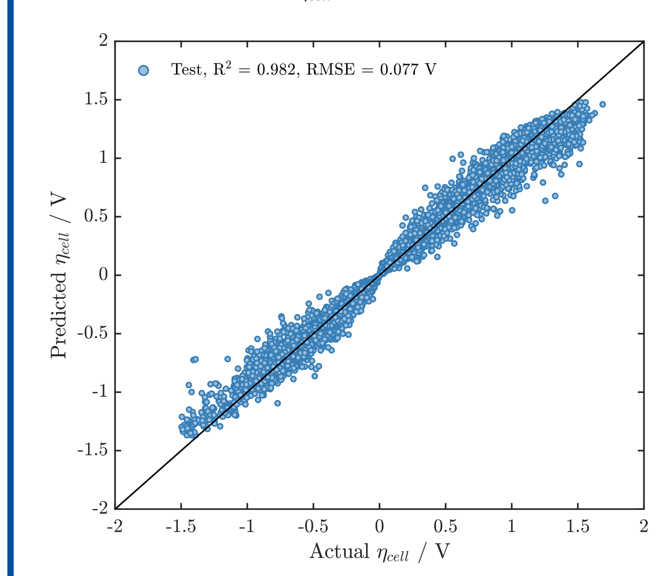
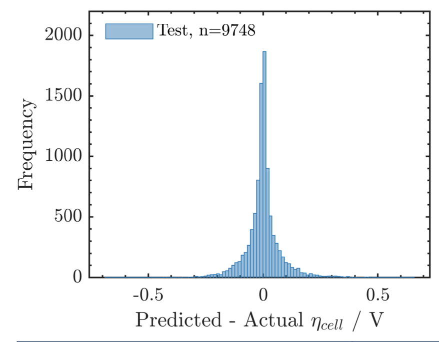


Figure 3. Distributions of operating conditions, electrolyte thickness, and cell overpotential with train and test expanding range -1.5 to 1.5 V.



- The test and OOB error rates converge to "asymptotic" minimum values and do not increase after the training error approaches zero, showing that RFR "does not overfit"
- Physics-based feature engineering significantly reduce the number of ML model inputs, speeding up training and execution times of ML model.
- Trained ML model shows high accuracy and has a run time of ~3 ms.





Time / h	1D PDE	ML model
Data (~200k)	336	0.17
Polarization curve (60 points)	0.10	5 x 10^{-5}

RFR for η_{cell}	
N_{pol}	195k
No. ξ_i	46
No. θ_i	29
No. features*	18
No. Trees*	100
Train Score	0.997
Test score	0.983
OOB score	0.981

Figure 4. Random forest regression analysis: error rates, final feature importances and correlations with η_{cell}, and training and testing results.

Validation against Multiphysics model and Literature data

PDE vs ML: Reversible solid oxide cell (SOC)

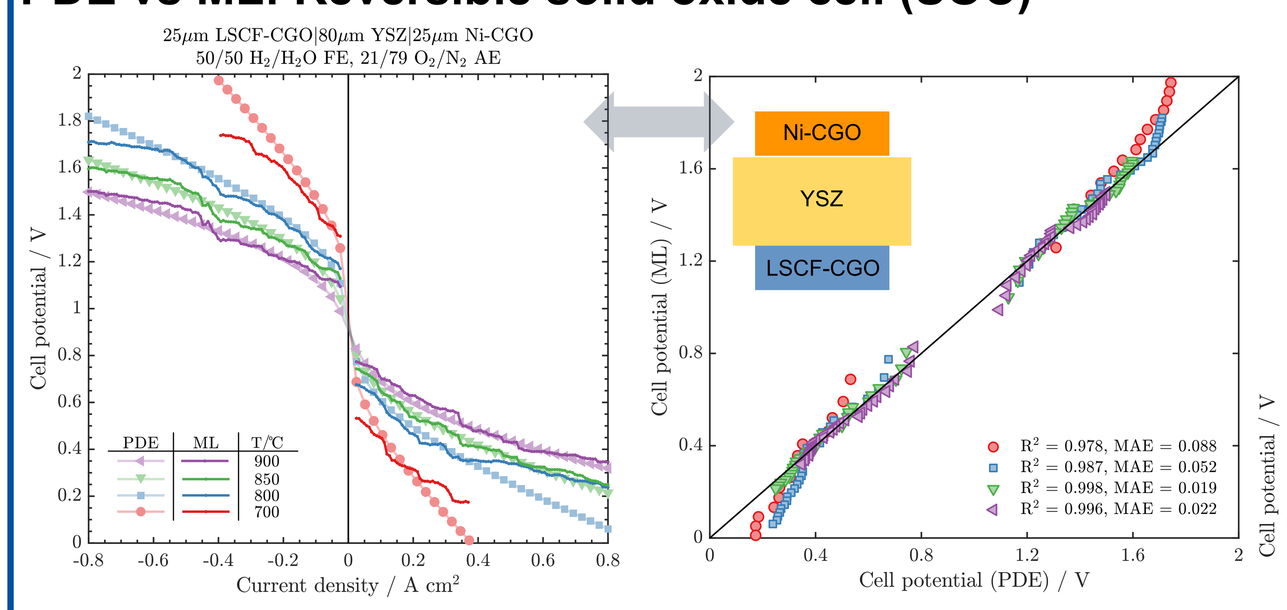
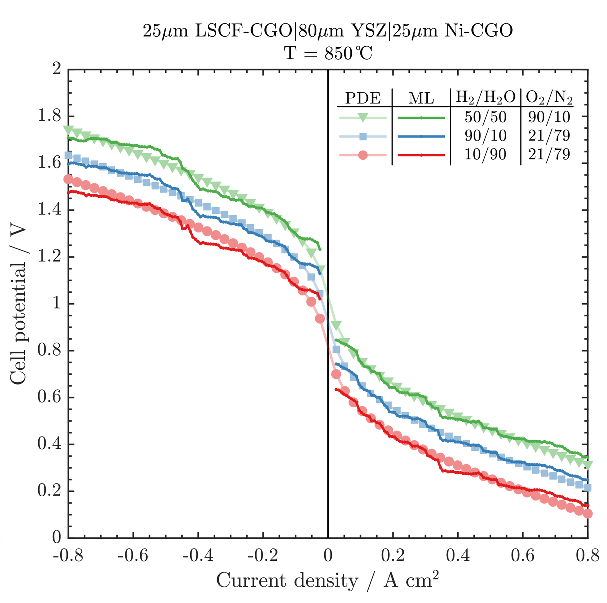


Figure 5. Comparing PDE vs ML model for different temperatures and same feed compositions.

ε (%)	d_p / μm	VF_{io} (%)	$A^V_{gas/ed}k^0$ / A m^{-3} $\lambda^V_{TPB}k^0$ / A m^{-3}	P /atm
30	0.4	50	10^7	1

Figure 6. Comparing PDE vs ML model for different feed compositions at a "well" trained temperature.



Literature data: Solid oxide fuel cell (SOFC)

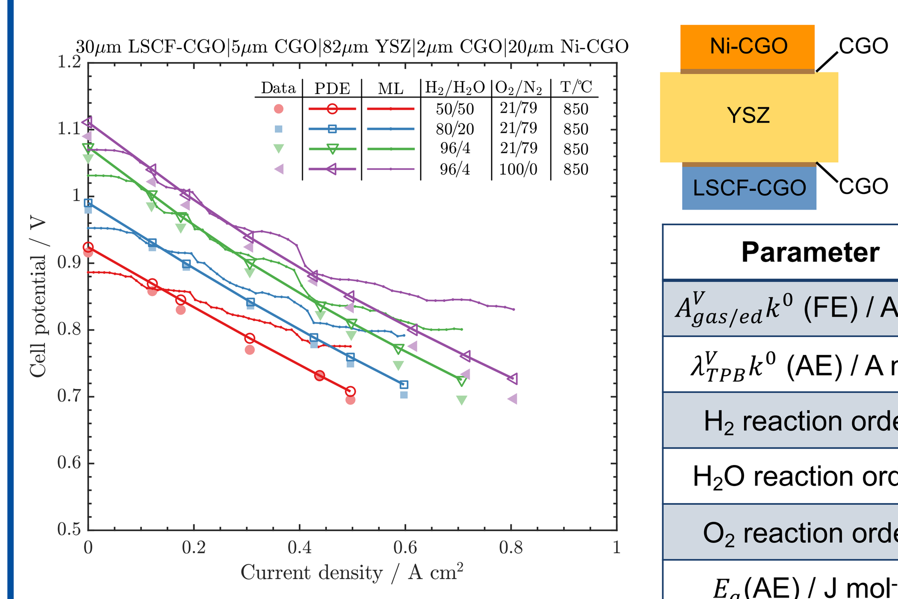


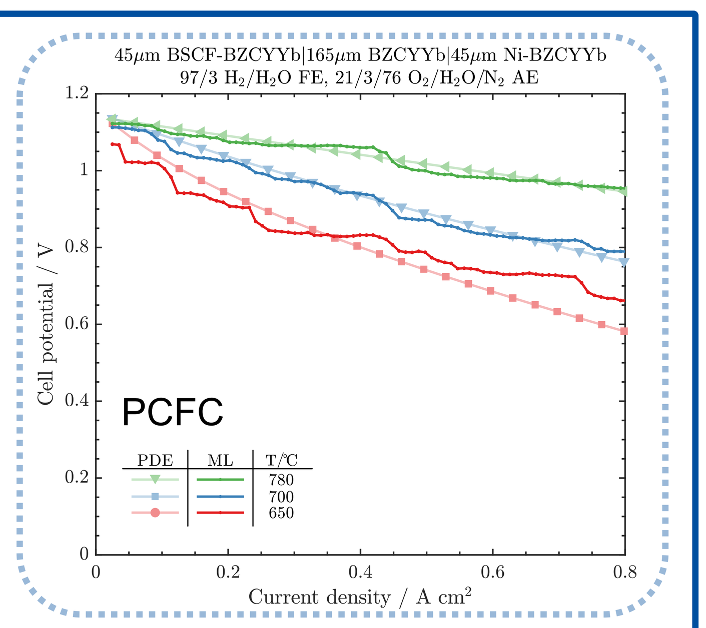
Figure 7. Comparing literature data [3] vs PDE and ML model. Fitting kinetic parameters for surface reactions in air and fuel electrode.

Parameter	Value
$A^V_{gas/ed}k^0$ (FE) / A m^{-3}	1.5x10^5
$\lambda^V_{TPB}k^0$ (AE) / A m^{-3}	447x10^7
H_2 reaction order	0.28
H_2O reaction order	0.0262
O_2 reaction order	0.199
E_a(AE) / J mol^{-1}	48688.47
E_a (FE) / J mol^{-1}	94355.81

- Reasonably good agreement for operation at higher temperatures.
- As temperature decreases and current density increases deviation from PDE overpotential solution is observed.
- ML model captures the effect of feed composition changes for "well" trained cases.
- Initial validation with literature data shows cell potential mismatch at high temperature
- Possibly, lack of enough polarisation data points for specific combination of physical parameters/features.

Next steps

- Expand data generated to capture the effect of individual physical parameters and their combined effect.
- Evaluation of other ML alternatives or their coupling with the RFR for better guess in regions with reduced polarisation points.
- Validation against extensive experimental data: ESC, ASC, MSC.
- Test methodology on other systems like PCFC →
- Coupling with hierarchical multiscale modelling framework to allow for lifetime simulation.



References:
1. Wehrle et al, ACS Environ. Au, 2022, 2, 42–64.
2. Wehrle et al, Chem. Ing. Tech., 2019, 91, No. 6, 833–842.
3. Padinjarethil et al, Front. Energy Res., 2021, 9:668964.