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YEARS 50 OF INNOVATION

Optimisation Methods for Solid Oxide Fuel Cell Design and Operation

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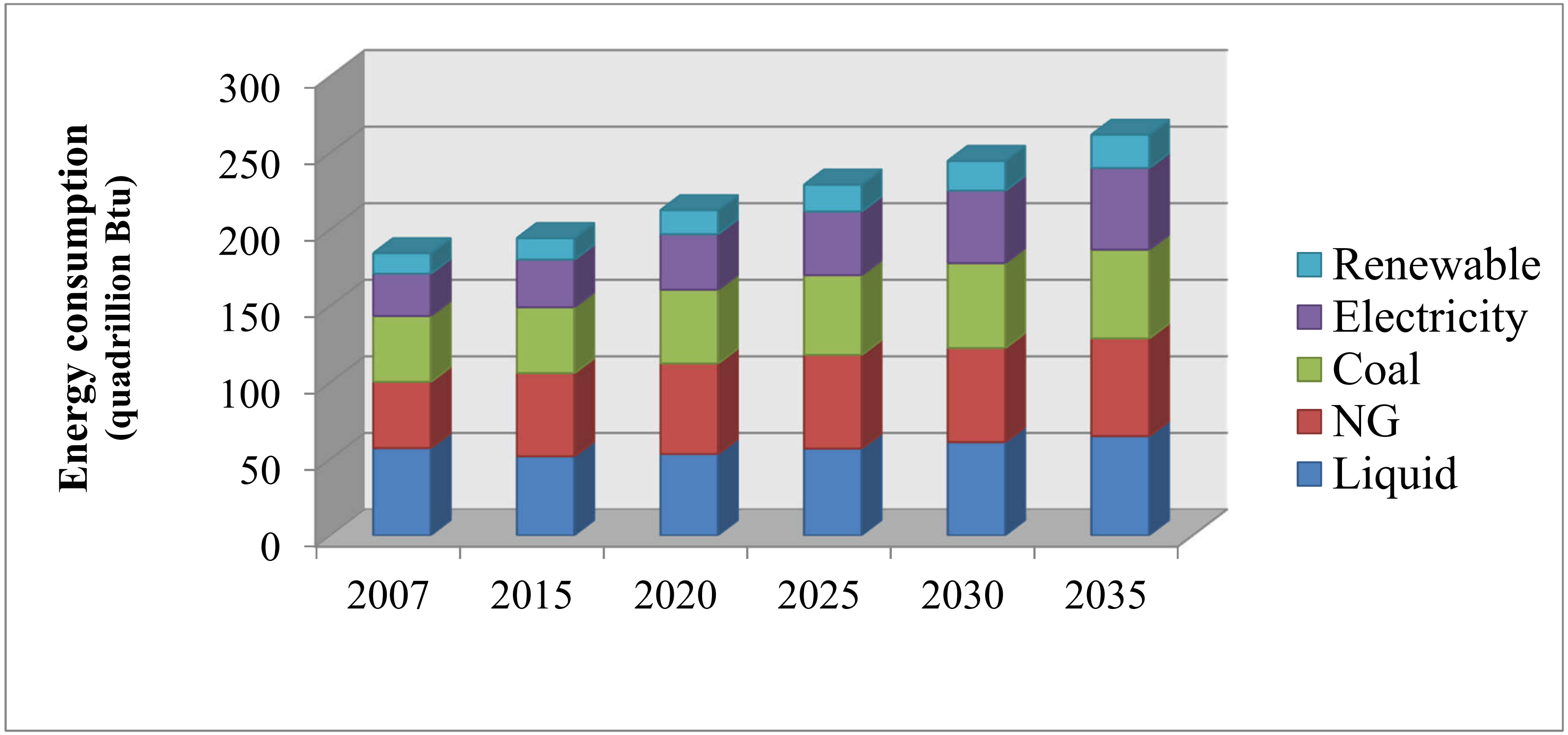
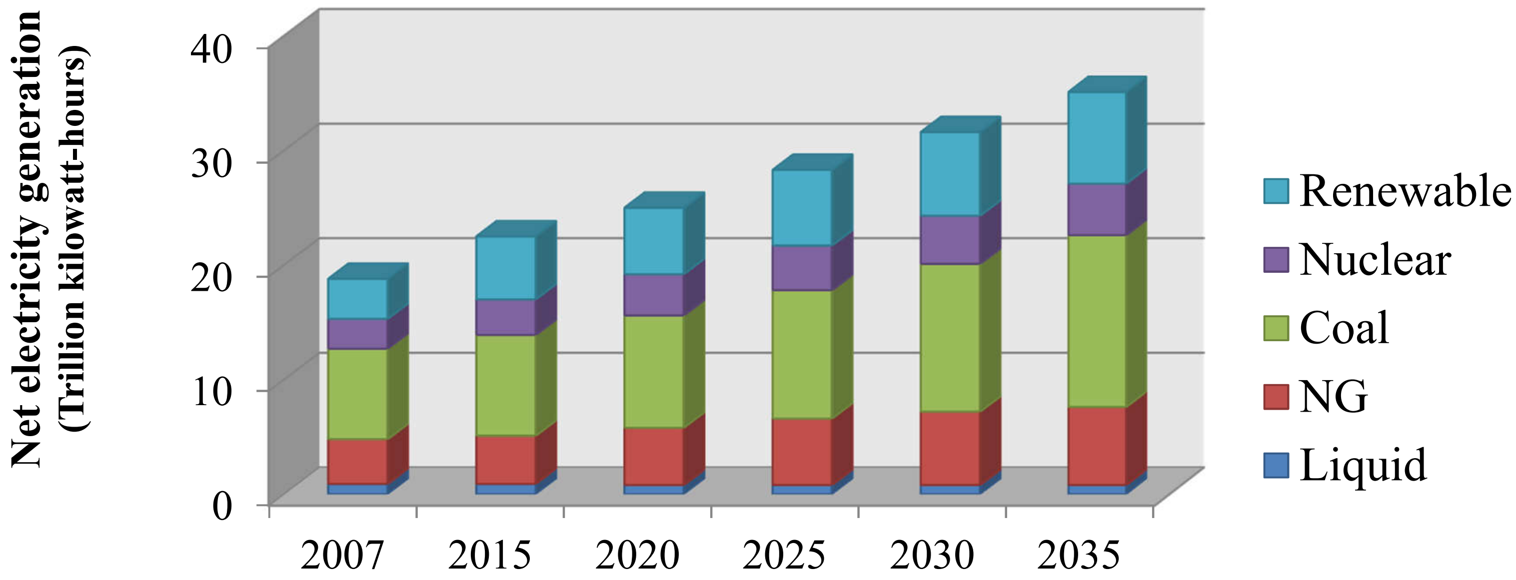
Projection for the world energy consumption by energy source

- World's energy consumption is increasing while electricity has the most rapid growth



Projection for the world net electricity generation by energy source

- Conventional fuels are expected to remain as the most important energy sources for electricity generation



Projection for the world net electricity generation by energy source

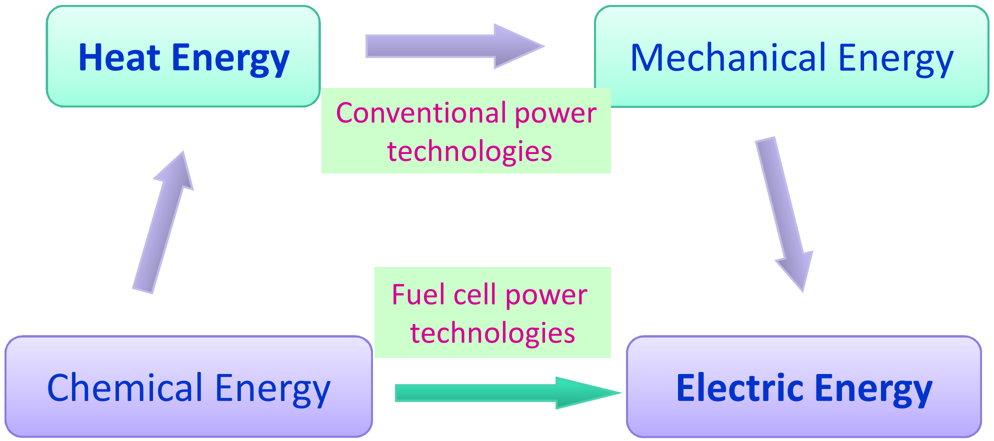
Research prospects for energy usage

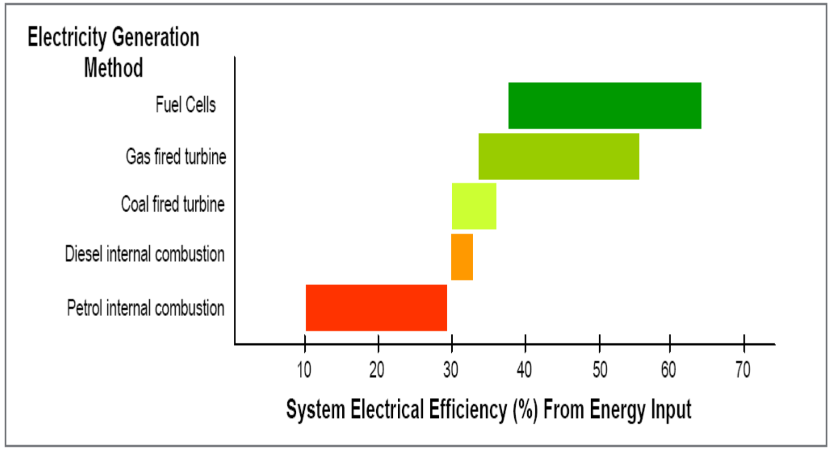
- The search is on...

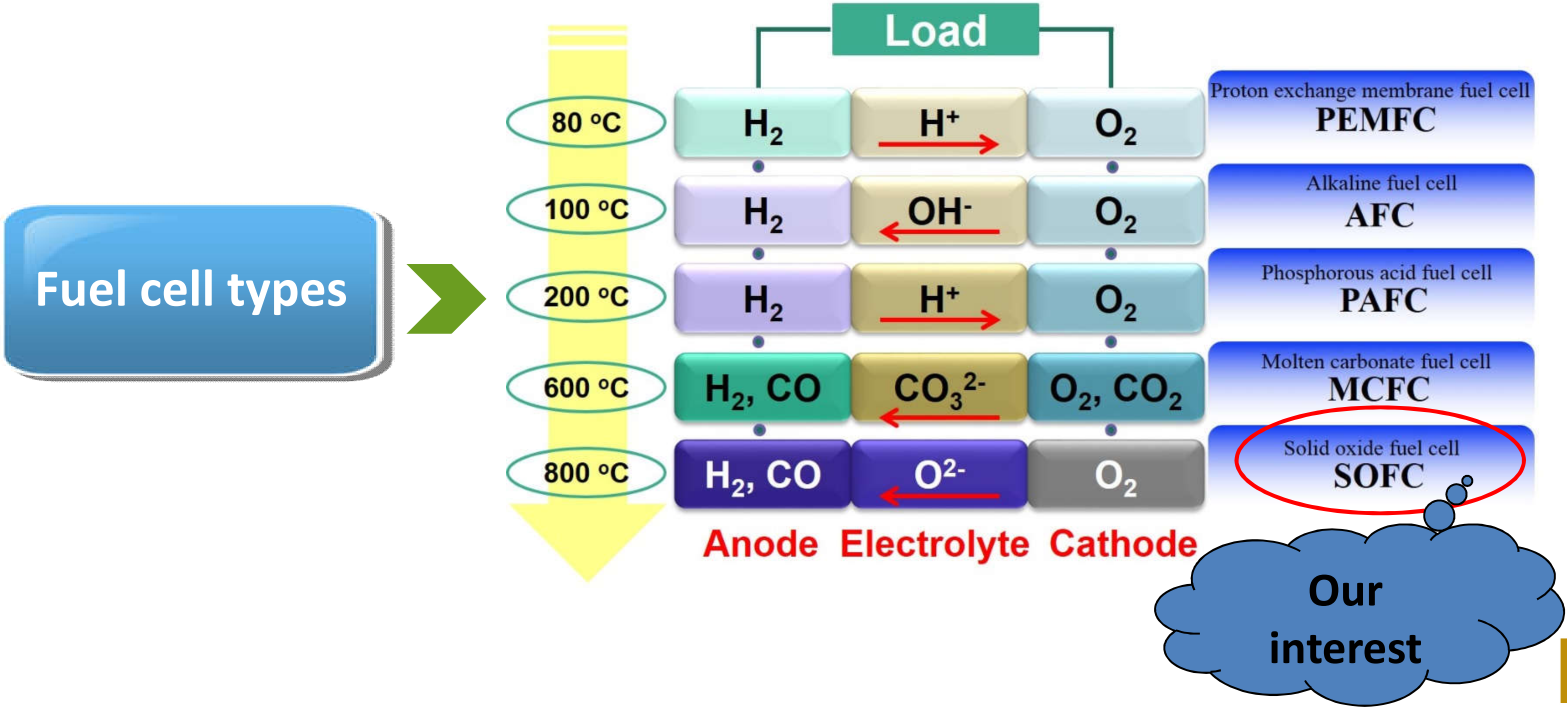
CFCL-Fuel cell test station

- For development of less polluting and more efficient energy conversion devices capable for sustainable consumption of different energy sources

A brief introduction to fuel cells







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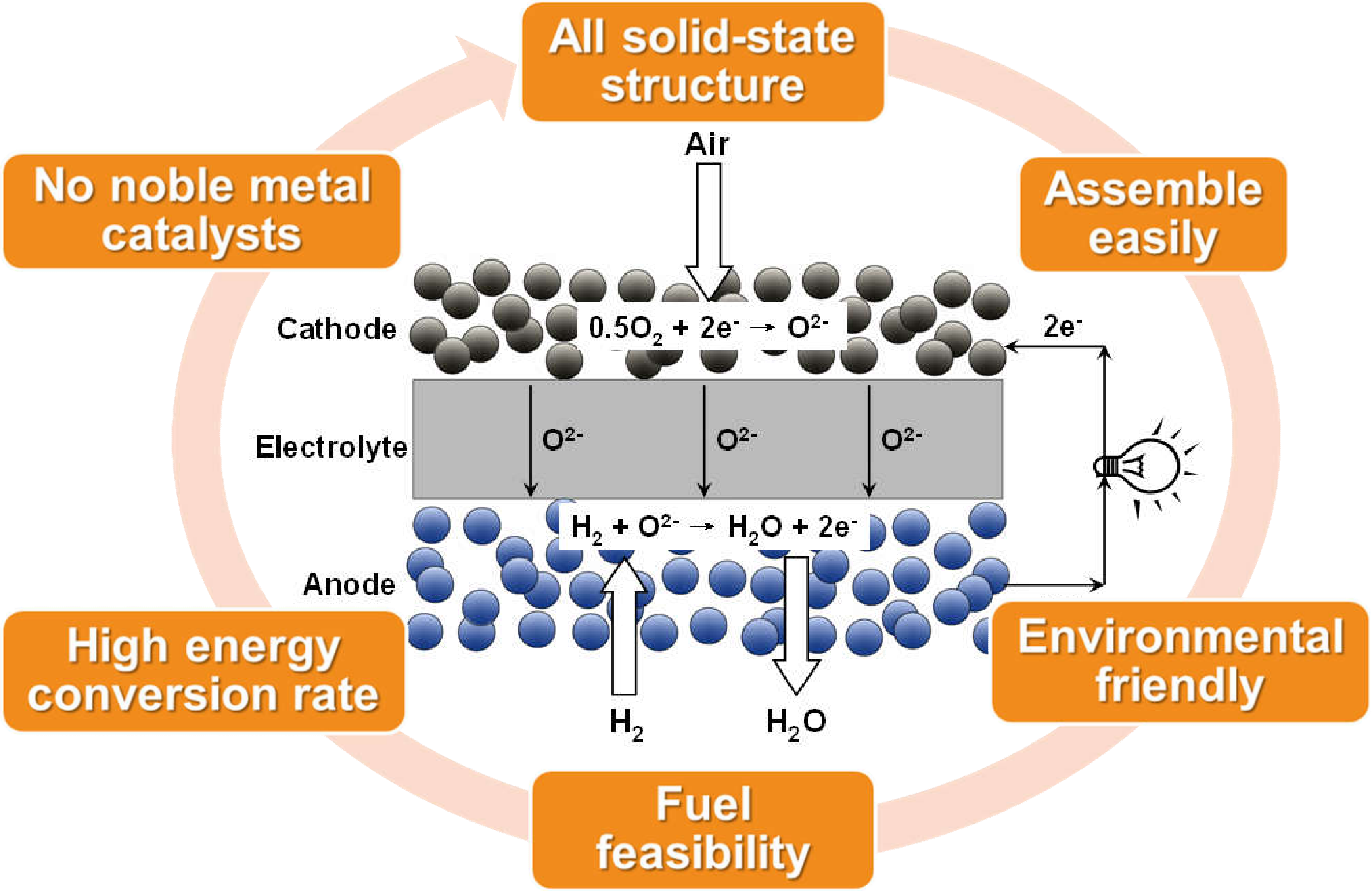
Solid oxide fuel cells (SOFCs)



Optimisation applications in SOFC

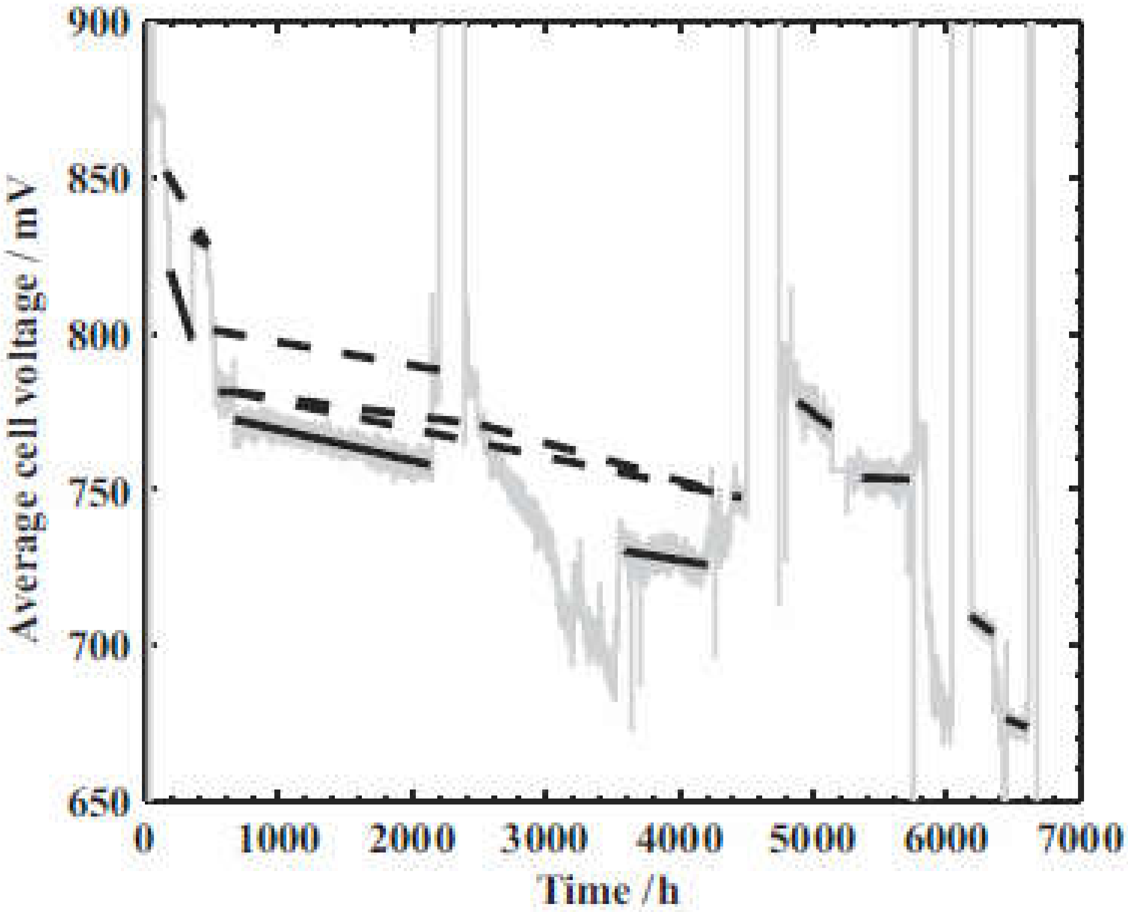
- Optimisation for thermal management
- Optimisation methods in model validation
- Electrode microstructure optimisation
- Energy and exergy optimisation

Optimisation for thermal management

Need for SOFC thermal management

- To maintain stack temperature at a certain level to obtain reasonable ion conductivity of the electrolyte
- To reduce thermal gradients in the stack to minimise thermal stresses
- For efficient heat integration of the system
- To maximise the overall efficiency of the system by devising a combined heat and power system
- To increase cell life by reducing cell degradation
- To avoid hot spots in the cell

Effect of cell degradation over time



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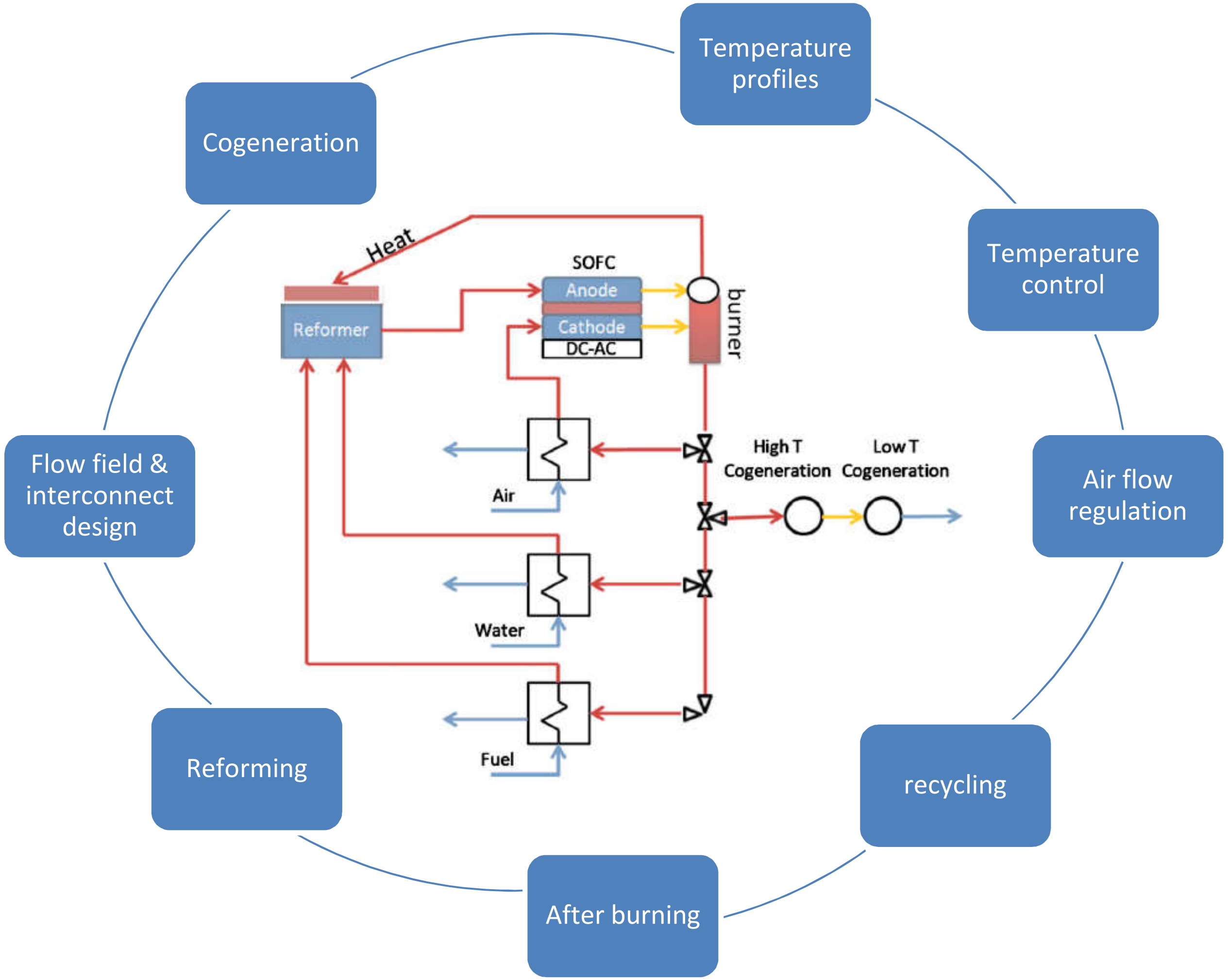
Aspects of SOFC thermal management



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Boundary level & dimensionality in SOFC multi-scale modelling

Electrode Level	Cell & stack level	System level
To explore the influences of the PEN structure on the reaction and transport processes	To investigate the cell & stack internal processes	Emphasize is focused on interaction of fuel cell with the other components of the system

- **Spatial variation is neglected (set of ODEs);**
- **Although the geometrical effects are neglected, micro processes or phenomena occurring within the PEN are studied in details;**
- **Less complexity;**
- **Computationally fast;**
- **Applicable for system level modelling, dynamic analysis and control system design.**

0D

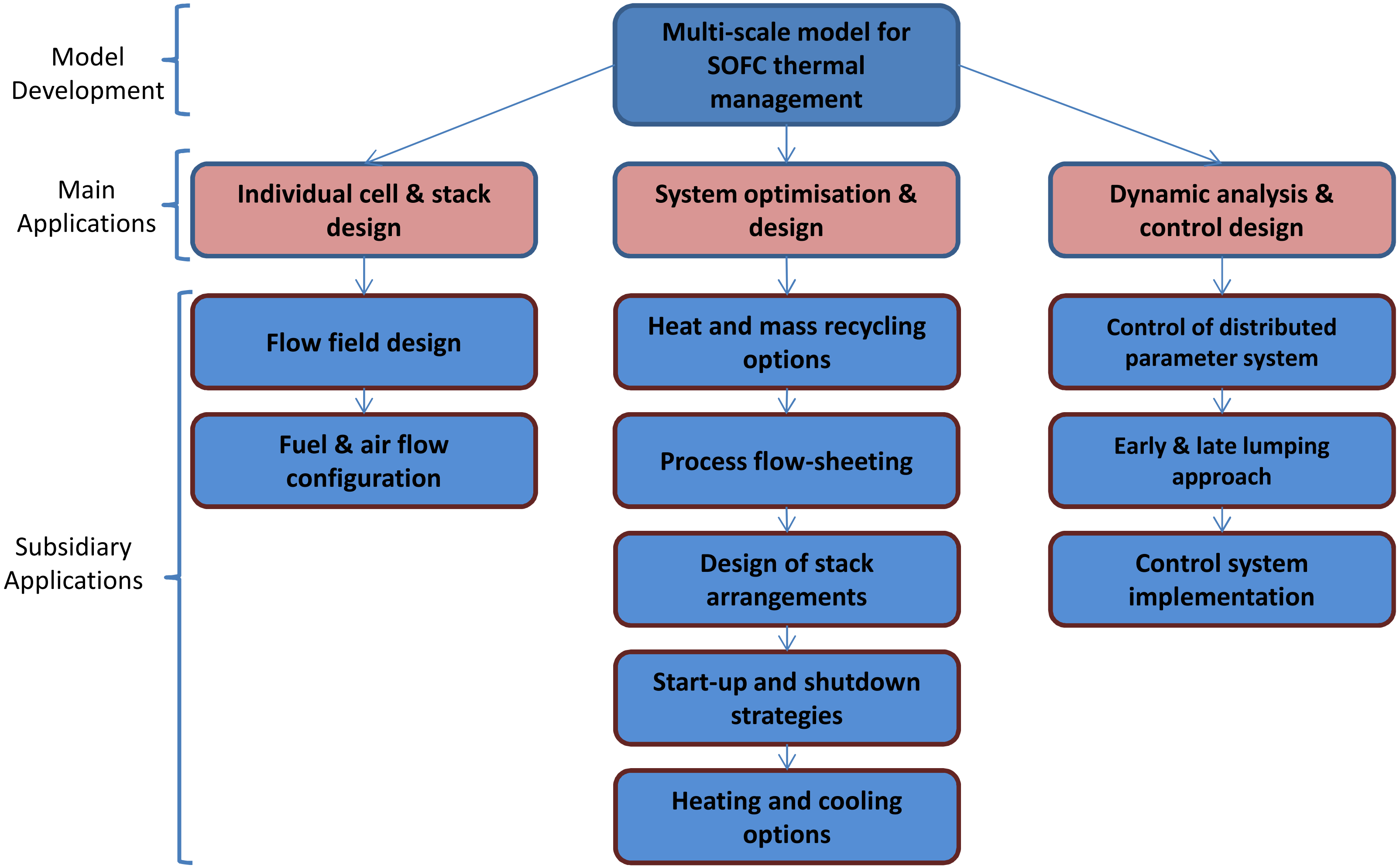
1D

2D

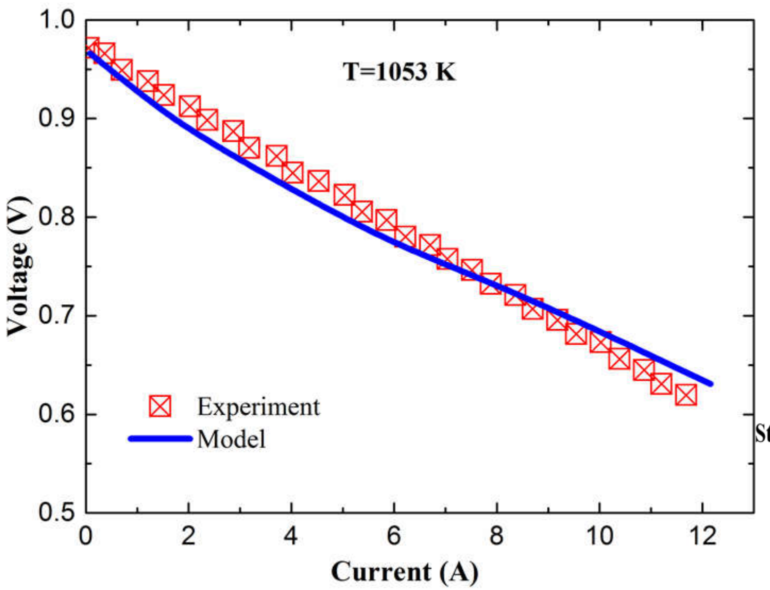
3D

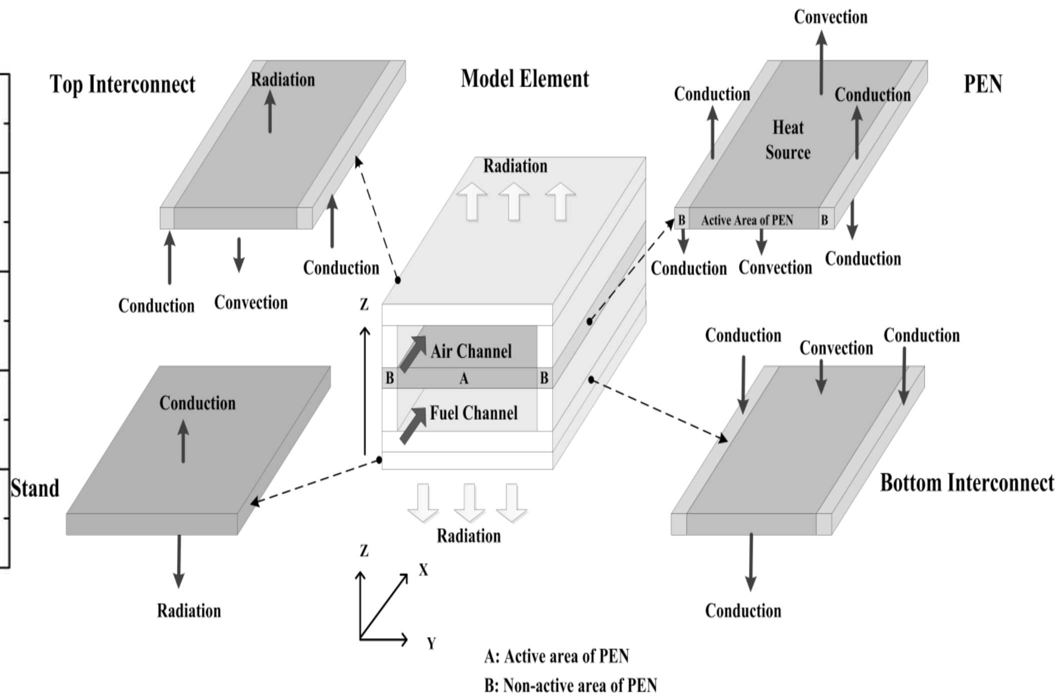
- **Spatial variation is considered (set of PDEs);**
- **Although micro processes are grossly simplified, the impact of the fuel and air flow configurations and different flow field design are taken into account;**
- **High complexity;**
- **Computationally slow;**
- **Applicable for the steady state performance analysis and flow field design of the cell/stack.**

Applications of a multi scale model for SOFC thermal management



Cell level multi-Layer modelling structure and validation



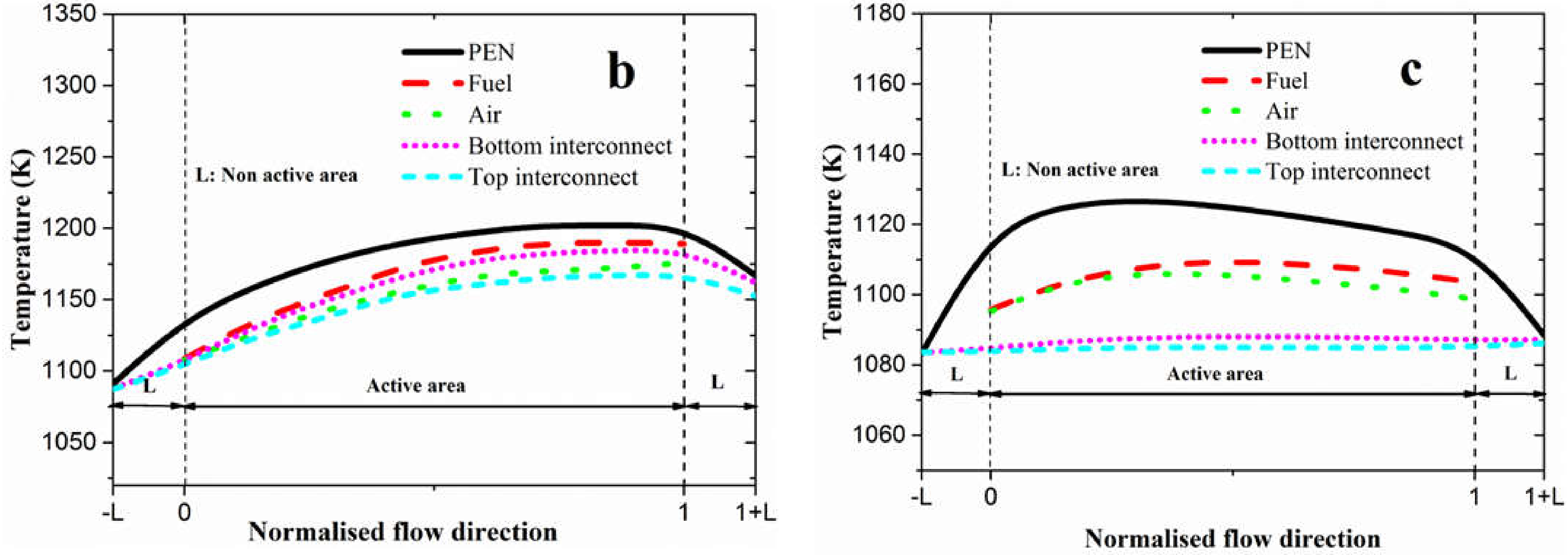


Four-layer modelling framework including stand, two interconnects, and PEN

Model Capabilities

- Detailed prediction of cell performance and thermal behaviour under both adiabatic and non-adiabatic conditions;
- Adiabatic condition is mostly consistent with cell's operation in stack (the marketable/commercial scale);
- Non-adiabatic condition is mostly consistent with cell's operation integrated with a furnace (laboratory scale);
- Detailed insights for temperature profiles in cells structure provides a design and thermal management tool;
- Model-based design of new tests is feasible

Temperature profiles in structural layers Adiabatic (left) and non-adiabatic(right)



System level modelling and simulation

Challenges:

→A black box/lump module is significantly numerically efficient;
BUT misses the main features of SOFC

→A distributed module accounts for detailed transport and reaction kinetics;
BUT causes serious complexity

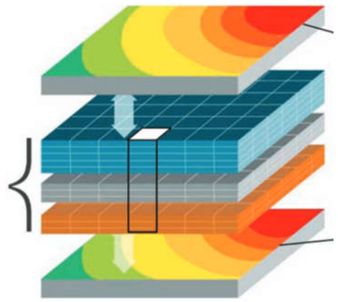
→More dimensions → more analysis feasible;
BUT more numerical facilities and proficiency needed

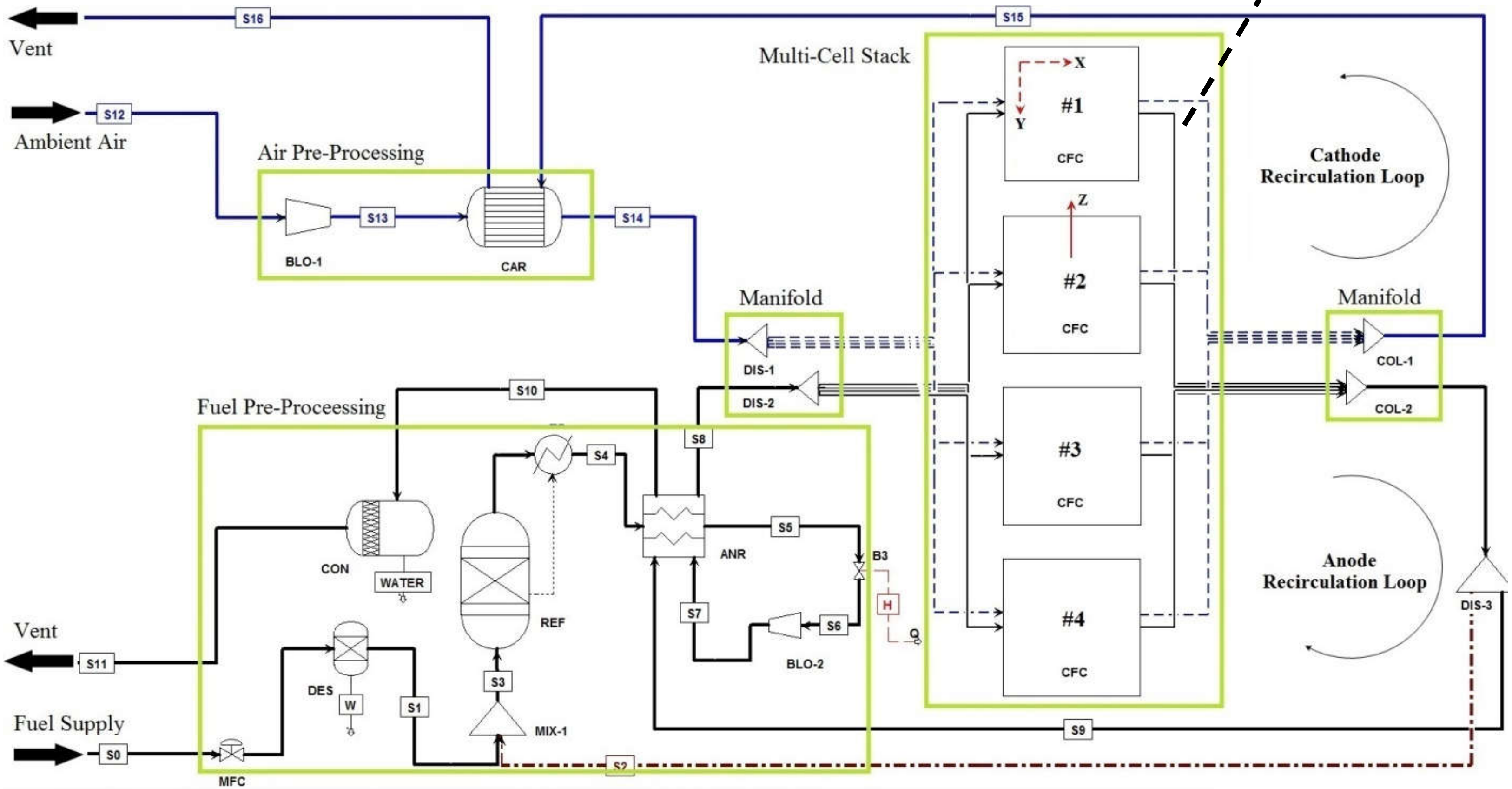
→A flowsheeting package is not a suitable tool for discretization purposes due to meshing difficulties;
BUT offers thermodynamics data bases and process analysis and optimization facilities;

Solution: Compromise

Target: A Detailed Stack in Flowsheeting Environment

Modelling and simulation outlook





ANR: Anode Recuperator **BLO:** Blower

CAR: Cathode Recuperator **COL:** Collector

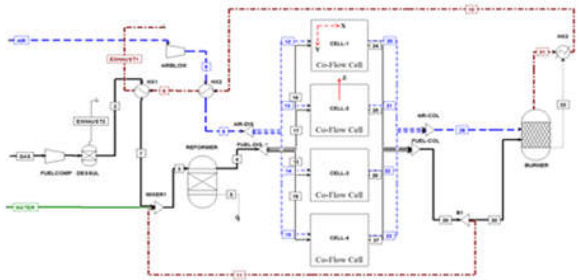
CON: Condenser **DES:** Desulpuriser

DIS: Distributor **MFC:** Mass Flow Controller

REF: Reformer **MIX:** Mixer

CFC: Co-Flow Cell

Modelling Platform



System Scale (1 m – 10 m)

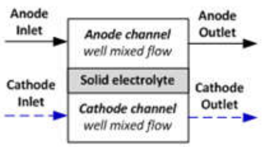
Typical Issues

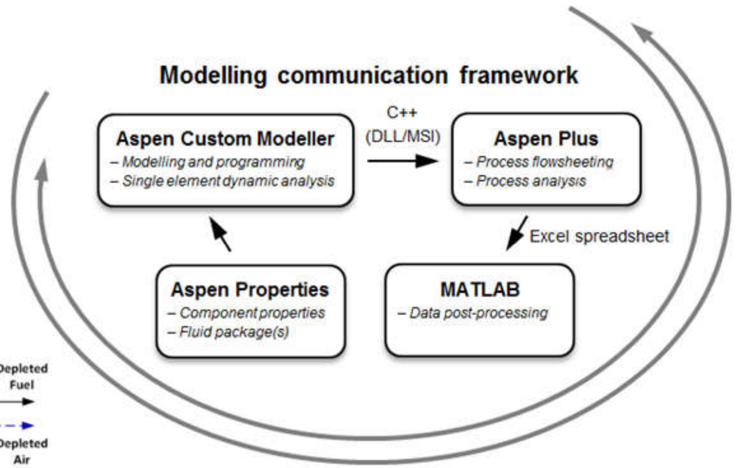
- Process optimisation
- Process dynamics and control
- Process commercialisation

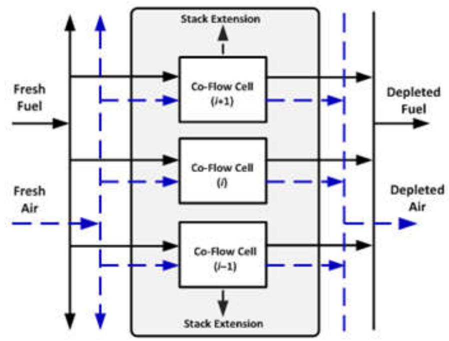
Compartment Scale (1 μm – 1 cm)

Typical Issues

- Reaction kinetics, heat and mass transfer
- Fuel conversion, carbon deposition
- Catalyst and material improvements



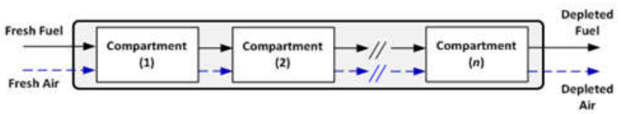




Channel Scale (1 cm – 10 cm)

Typical Issues

- Fuel and air flow configuration
- Fluid flow regimes
- Residence time distributions



Stack Scale (10 cm – 1 m)

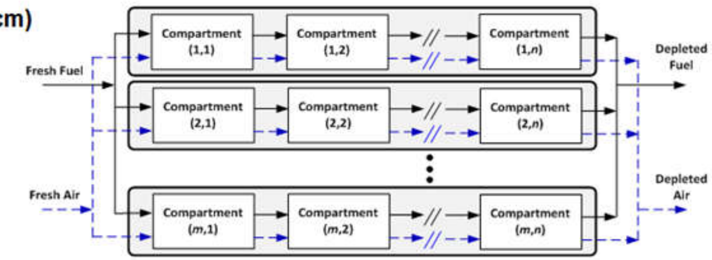
Typical Issues

- Stack energy integration
- Process steadiness
- Start-up and shut-down dynamics

Cell Scale (1 cm – 10 cm)

Typical Issues

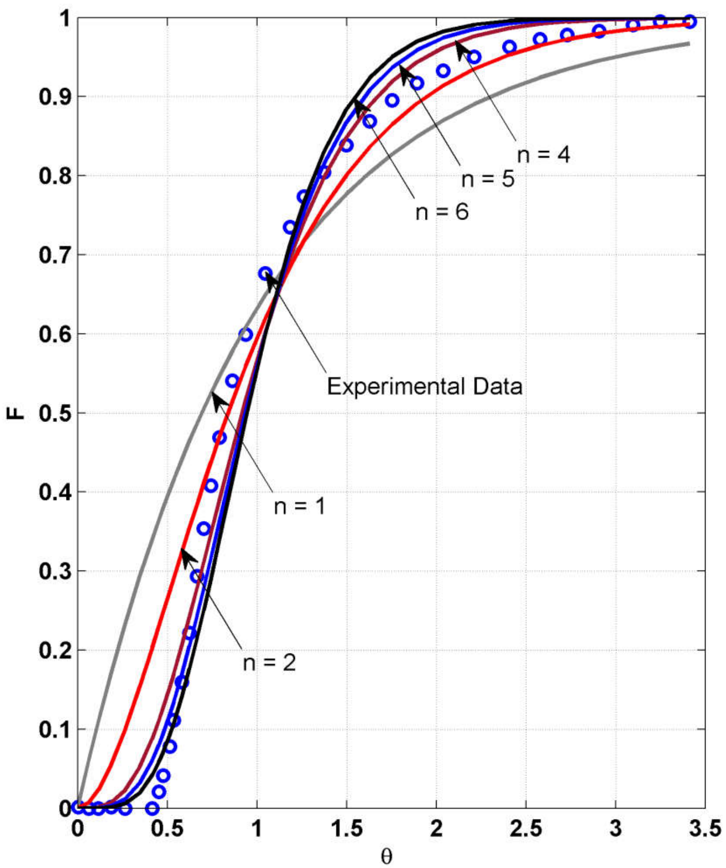
- Thermal management
- Cell life durability
- Dynamic cell behaviour



Model capabilities

- Prediction of SOFC performance at different scales and with adjustable details(0-3D);
- System level modelling, flowsheeting, analysis, and design;
- Optimization capability through both operating variable manipulation and process flow diagram improvement;
- Potential room for dynamic and control research at system level;
- Utilization of well-stablished components data bases and thermodynamical packages;
- Establishment of a modular modelling library customised for electrochemical reactors such as SOFC, MCFC, ...
- Fuel processing analysis without further programming/modelling needs;

Estimation of number of compartments in series based on reactor's Residence Time Distribution

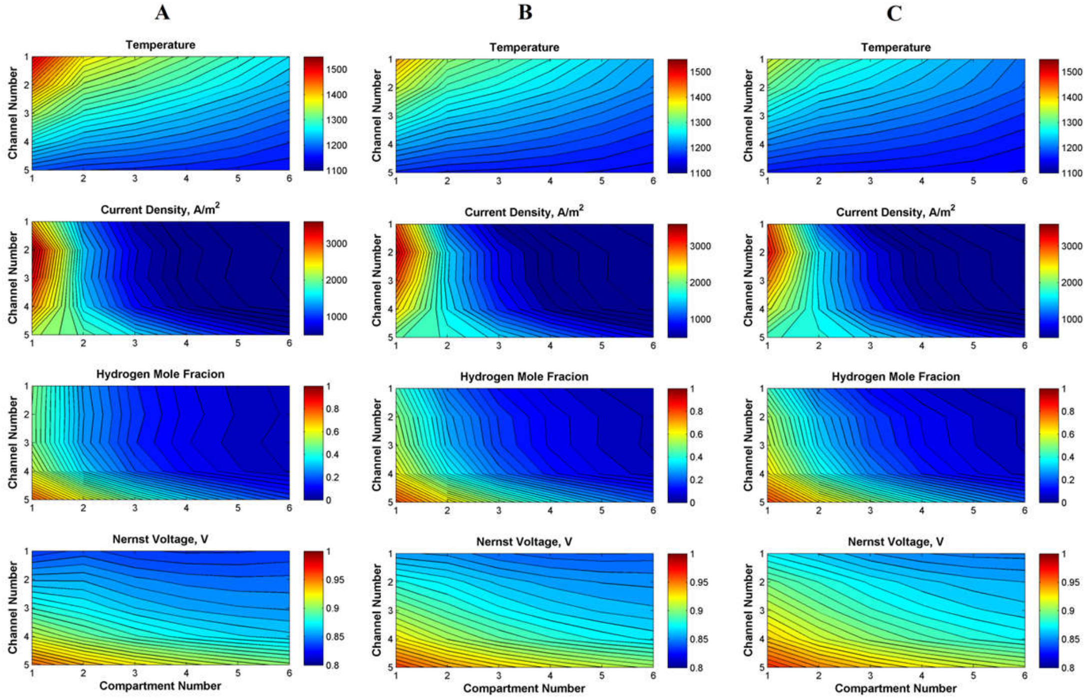


Circles:
Experimental RTD data (Krewer et al. (2004))

Solid lines:
Prediction based on n compartments

$$F = 1 - e^{-n\theta} \sum_{i=1}^{n} \frac{(n\theta)^{i-1}}{(i-1)!}$$

Sensitivity Analysis at Cell Scale: Air Flow Rate Impact

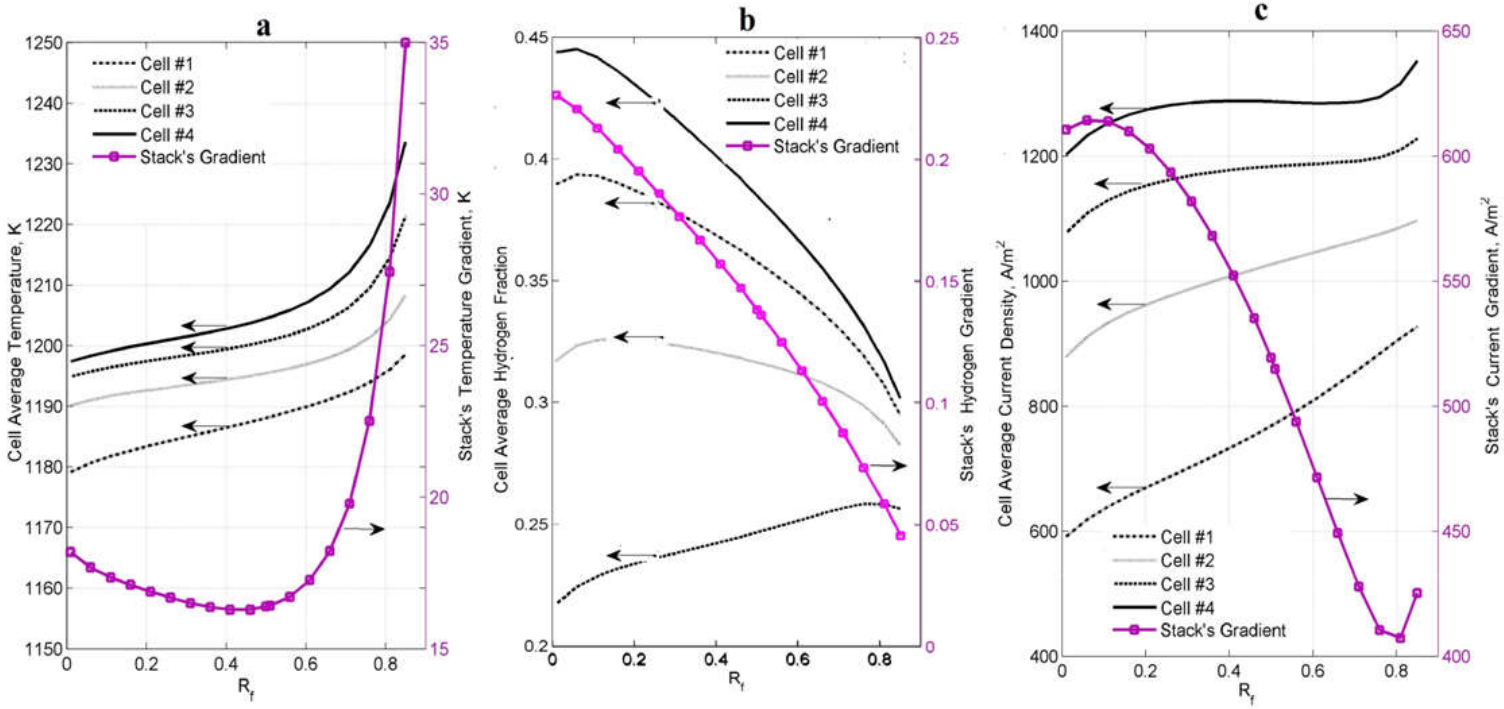


Higher air flow rate results in

- more homogeneity in cell's distributed variables
- Lower average temperature and supressing the electrochemical reaction/current generation;
- Higher pressure drop/costs

Variable	A (Air flow rate 20% lower than base case)	B (Base case air flow rate)	C (Air flow rate 20% higher than base case)
Average Temperature, K	1265	1235	1215
Temperature Coefficient of Variation	0.0663	0.0522	0.0432
Average Current Density, A/m^2	1113	1108	1099
Current Density Coefficient of Variation	0.8854	0.8482	0.8119
Average Nernst Voltage, V	0.873	0.883	0.891
Average Hydrogen Mole Fraction	0.25	0.26	0.27
Fuel Utilization	0.91	0.90	0.89

Stacking direction gradients as a function of anode off-gas recycle (AOGR)

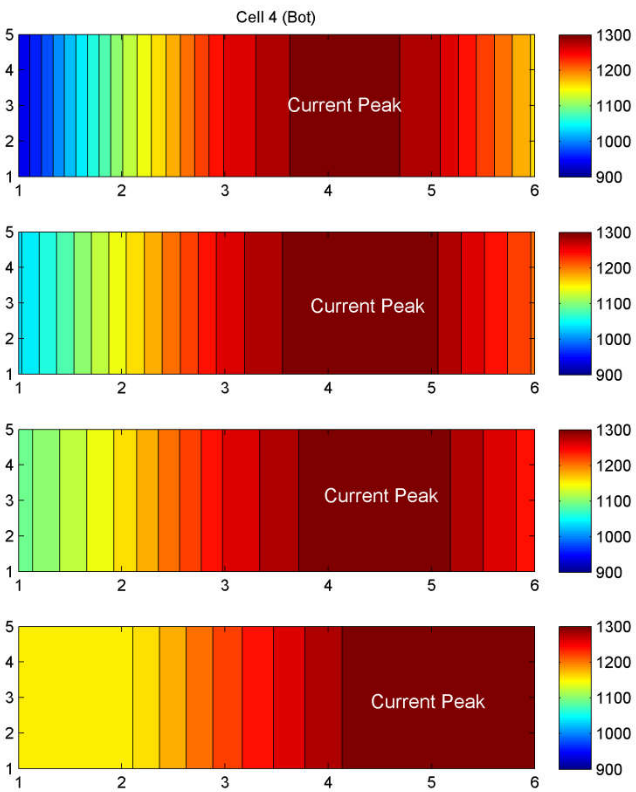


- AOGR rate has different effect on cells in stack depending on their fuel share;
- AOGR influences the gradient in stacking direction making room for optimization works for thermal management
- AOGR should be limited to due sizing and cost issues.

Counters for single cell unit and multi-cell stack under varying AOGR

AOGR

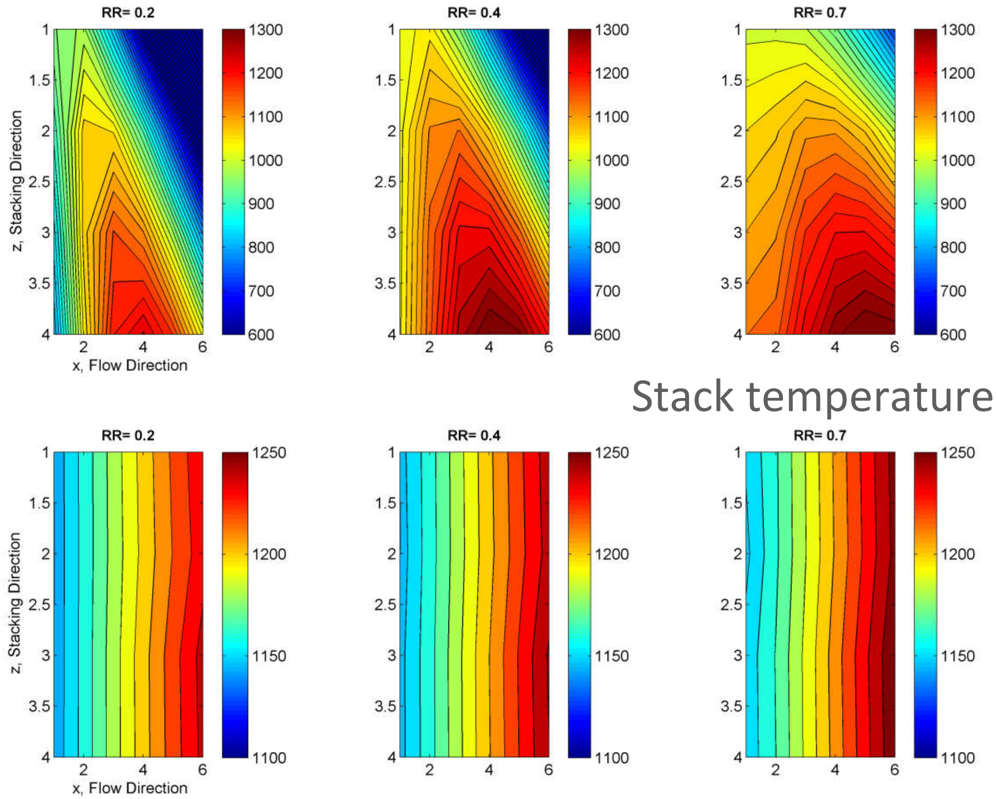
Single cell current



Flow direction

AOGR

Stack current



Stack temperature

Flow direction

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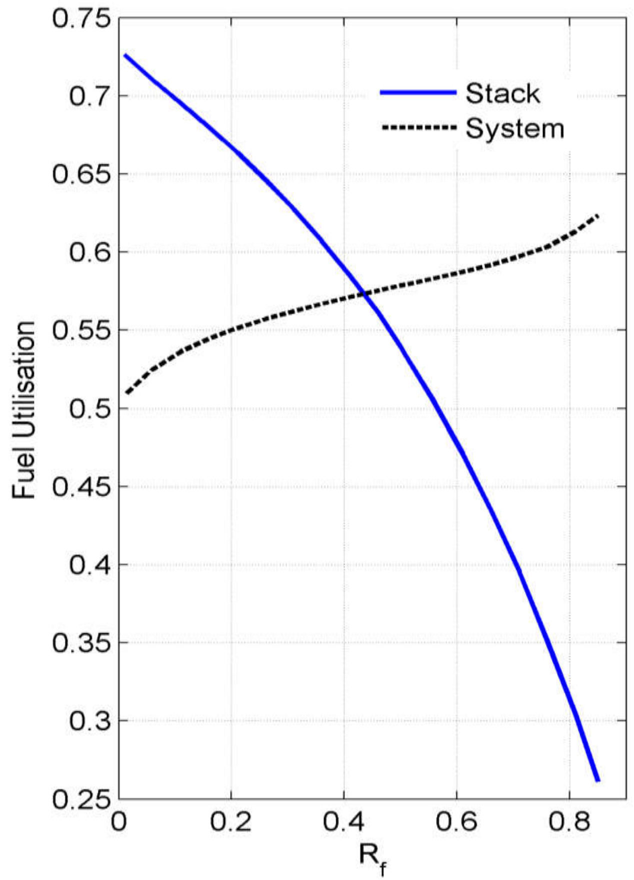
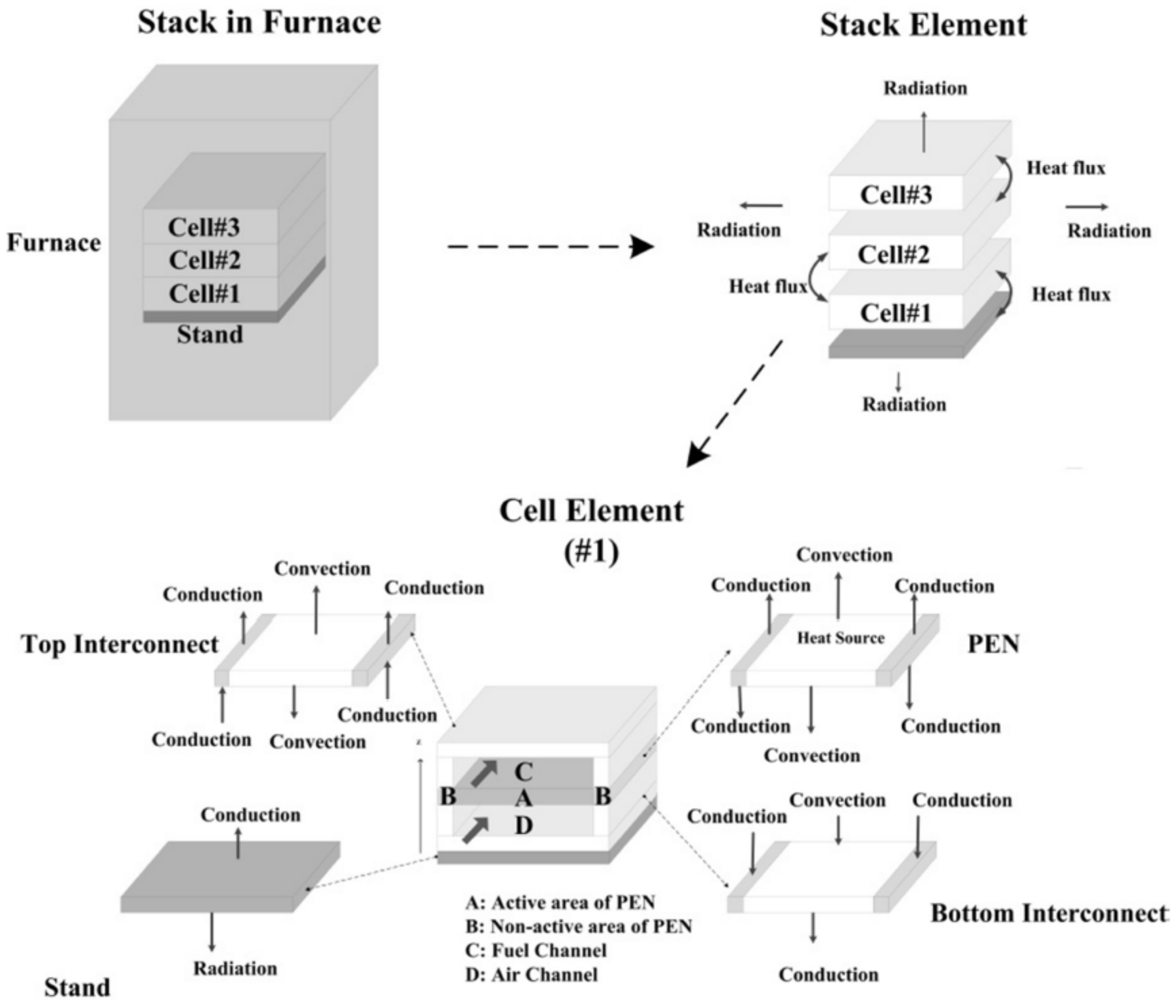
Process performance: Effect of recycle fraction on stack and system fuel utilisation

- AOGR reduces the fuel utilization in stack while increases that for whole system;
- This is technically desired because overall efficiency will be improved while stack/cell fuel starvation and hot spot formation can be avoided;
- Optimization of AOGR must be conducted through a detailed stack model integrated in a system level model such as this work.
- This optimization task is certainly a multi- objective one that leads in a so called “effective operation” not “most optimum operation” as it ultimately results in a compromised strategy.



3 cell short-stack for multi-objective optimisation



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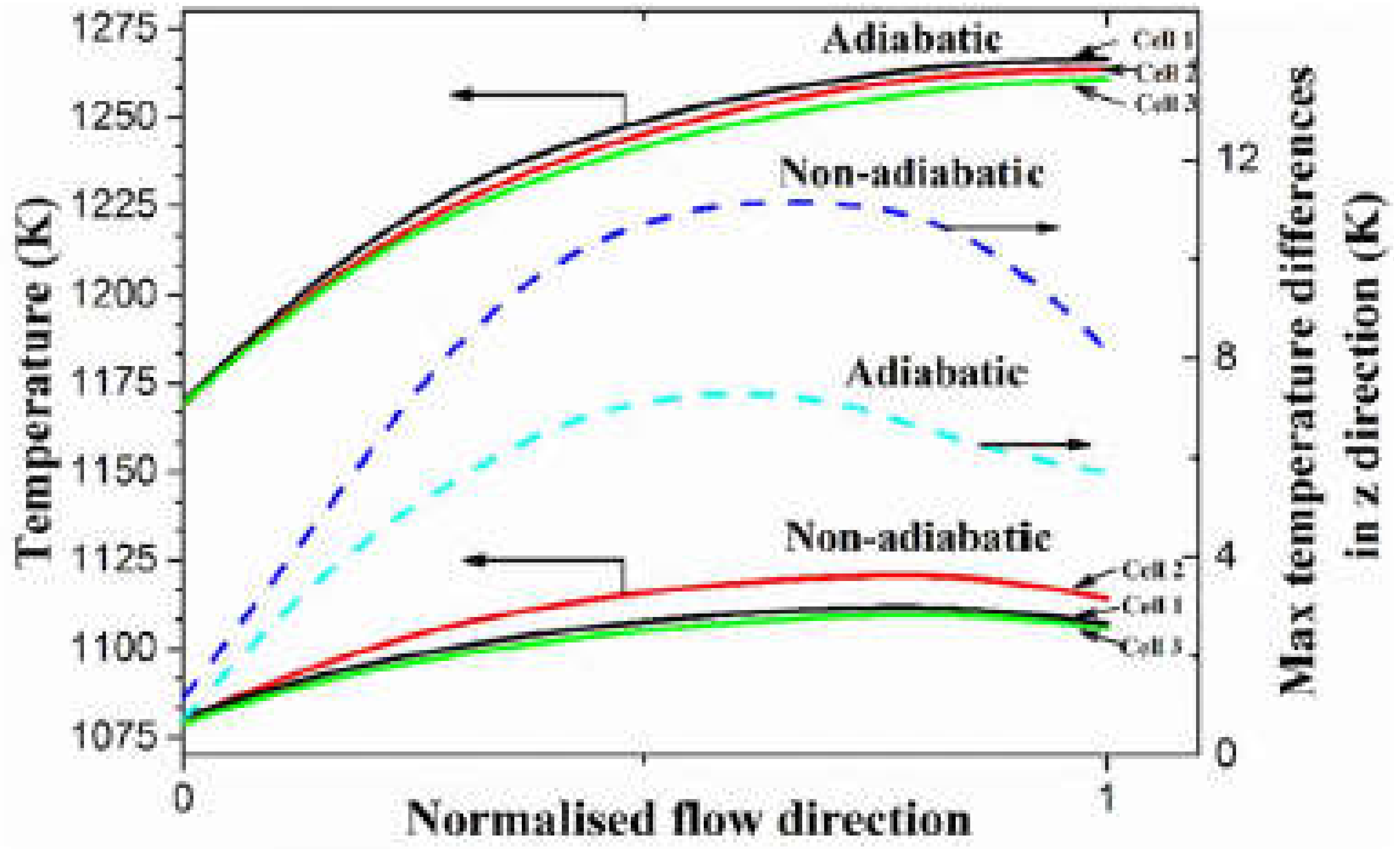
Temperature and efficiency calculation

equation	comment
$dT = \frac{\partial T}{\partial x}dx + \frac{\partial T}{\partial z}dz$	stack's total temperature gradient
$\bar{T}^{\text{stack}} = \frac{1}{3}\sum_{i=1}^{3}\bar{T}^{\text{cell},i}$	stack's average temperature
$\bar{T}^{\text{cell}} = \frac{1}{n}\sum_{i=1}^{n}T^{i}$	cell's average temperature
$\left(\frac{\partial T}{\partial x}\right)^{\text{cell}} \cong \frac{\Delta T^{\max}}{L_x}$	cell's temperature gradient in x direction
$\left(\frac{\partial T}{\partial x}\right)^{\text{stack}} = \frac{1}{3}\sum_{i=1}^{3}\left(\frac{\partial T}{\partial x}\right)^{\text{cell},i}$	stack's average temperature gradient in x direction
$\left(\frac{\partial T}{\partial z}\right)^{\text{stack}} \cong \frac{\Delta T^{\max}}{L_z}$	stack's temperature gradient in z direction

$$\text{Eff}_{\text{el}} = \frac{P_{\text{net}}}{F_{\text{H}_2,\text{in}}\text{LHV}_{\text{H}_2}}$$

Temperature profiles and maximum temperature differences in the stack



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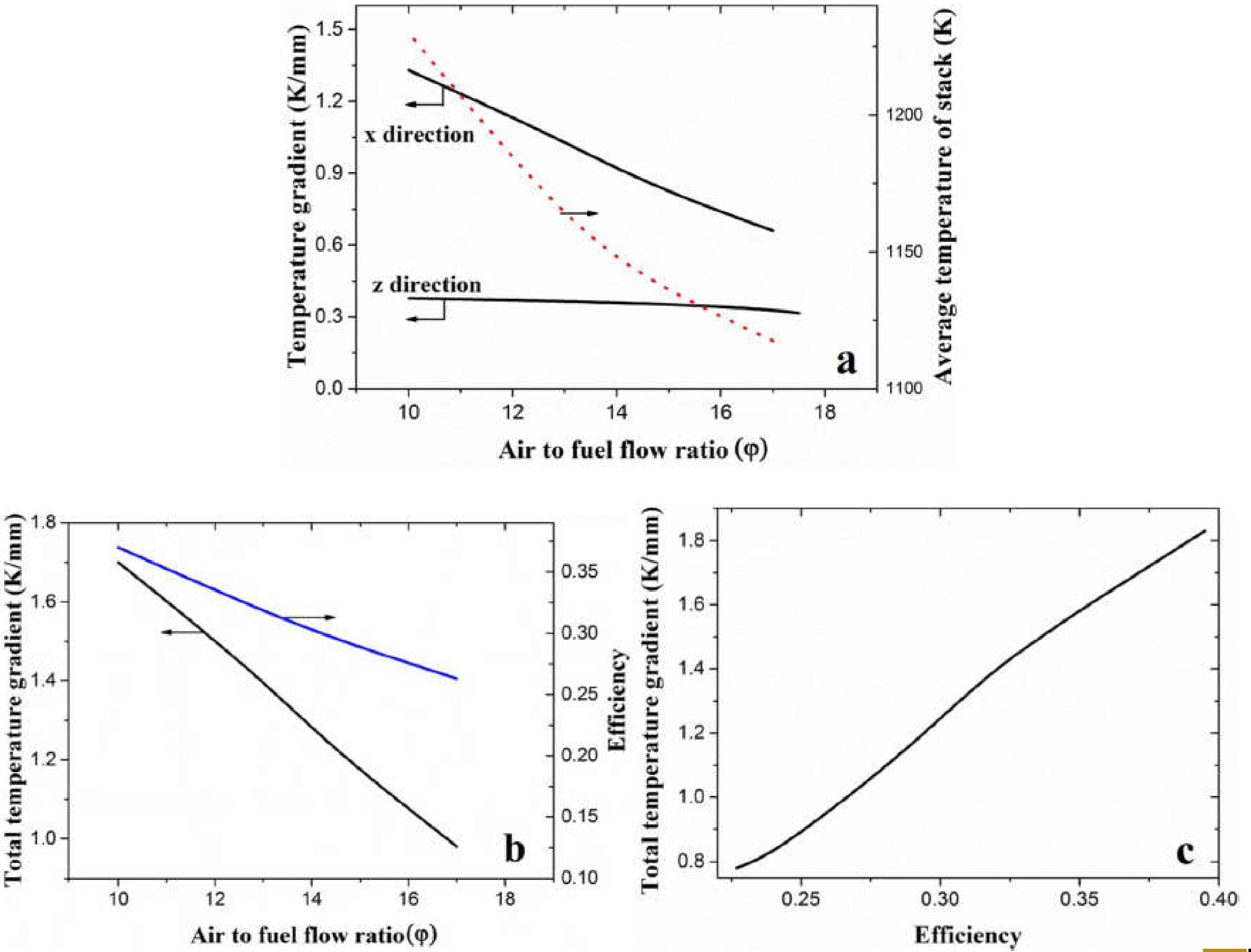
Strategies for gradient suppression

- Excess air flow
- Adjusting the temperature differences between the two inlet gas streams
- Utilisation of oxygen enriched air

Effect of excess air flow on temperature gradient and efficiency



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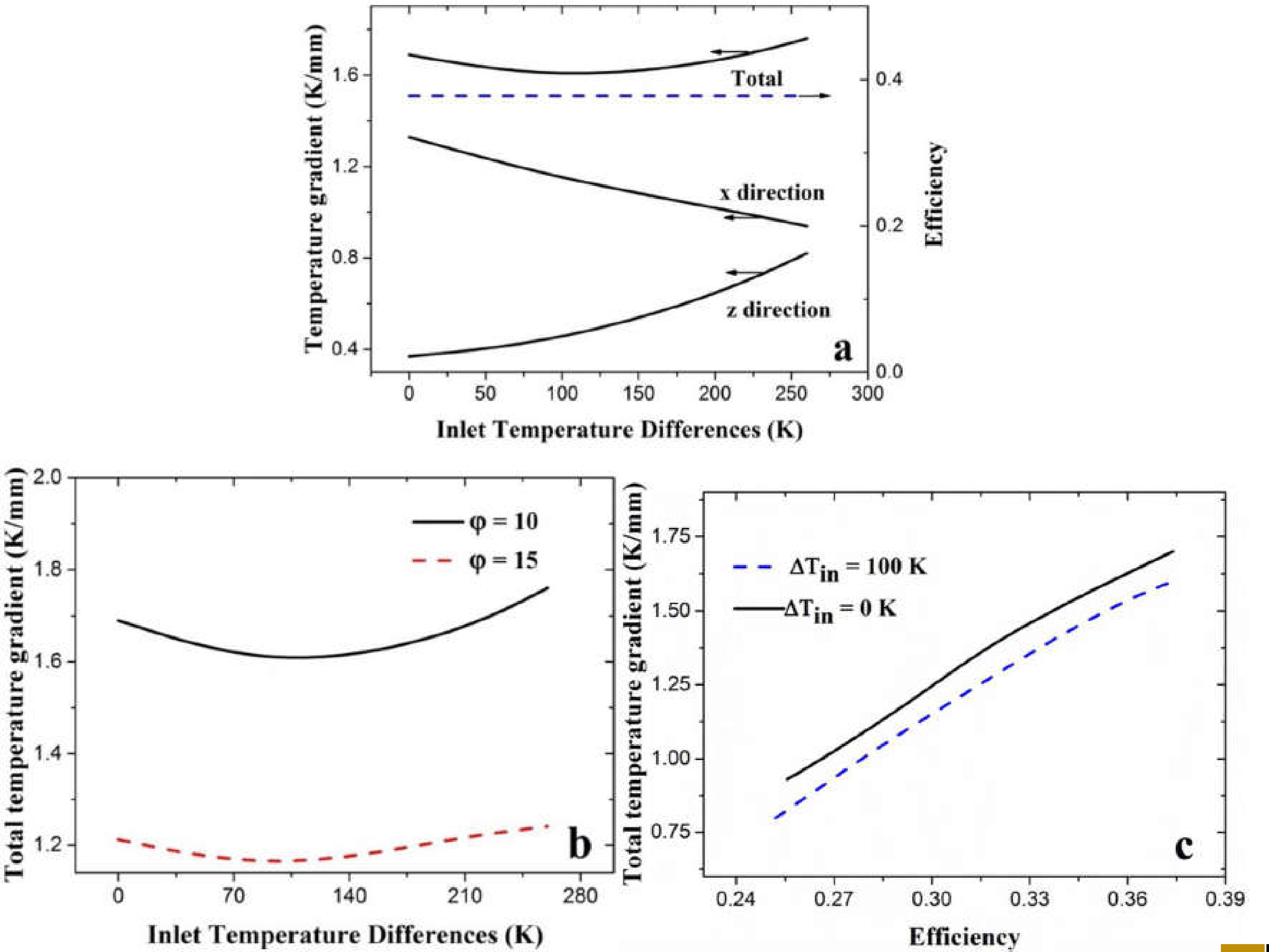
Effect of inlet temperature difference on temperature gradient and efficiency



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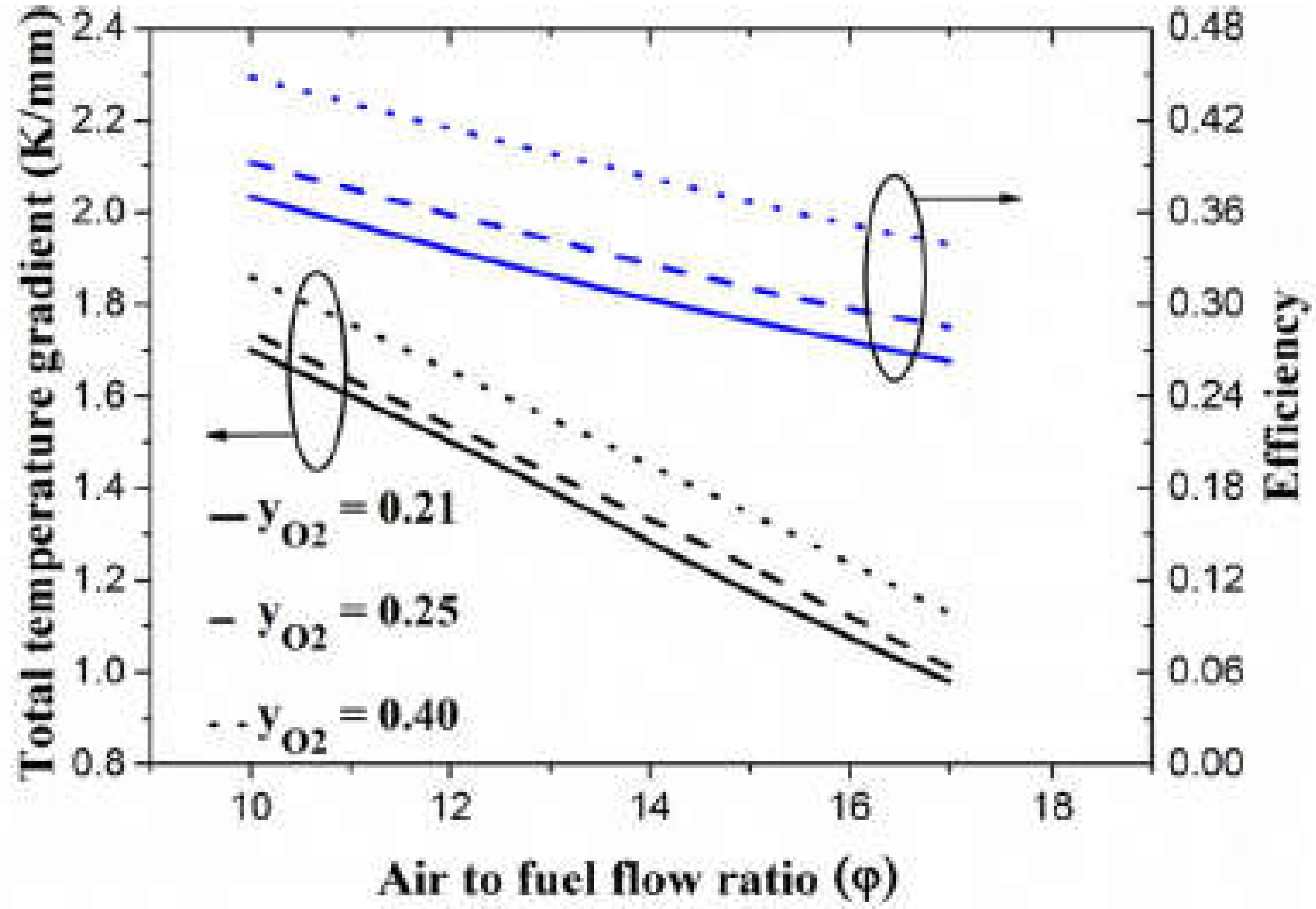
Oxygen concentration effect on temperature gradient and efficiency



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Multi-objective optimisation

Objective function

$$F_{ob} = \omega F_{\Delta T}(s) + (1 - \omega) F_{Eff}(s) \qquad \omega \in [0,1]$$

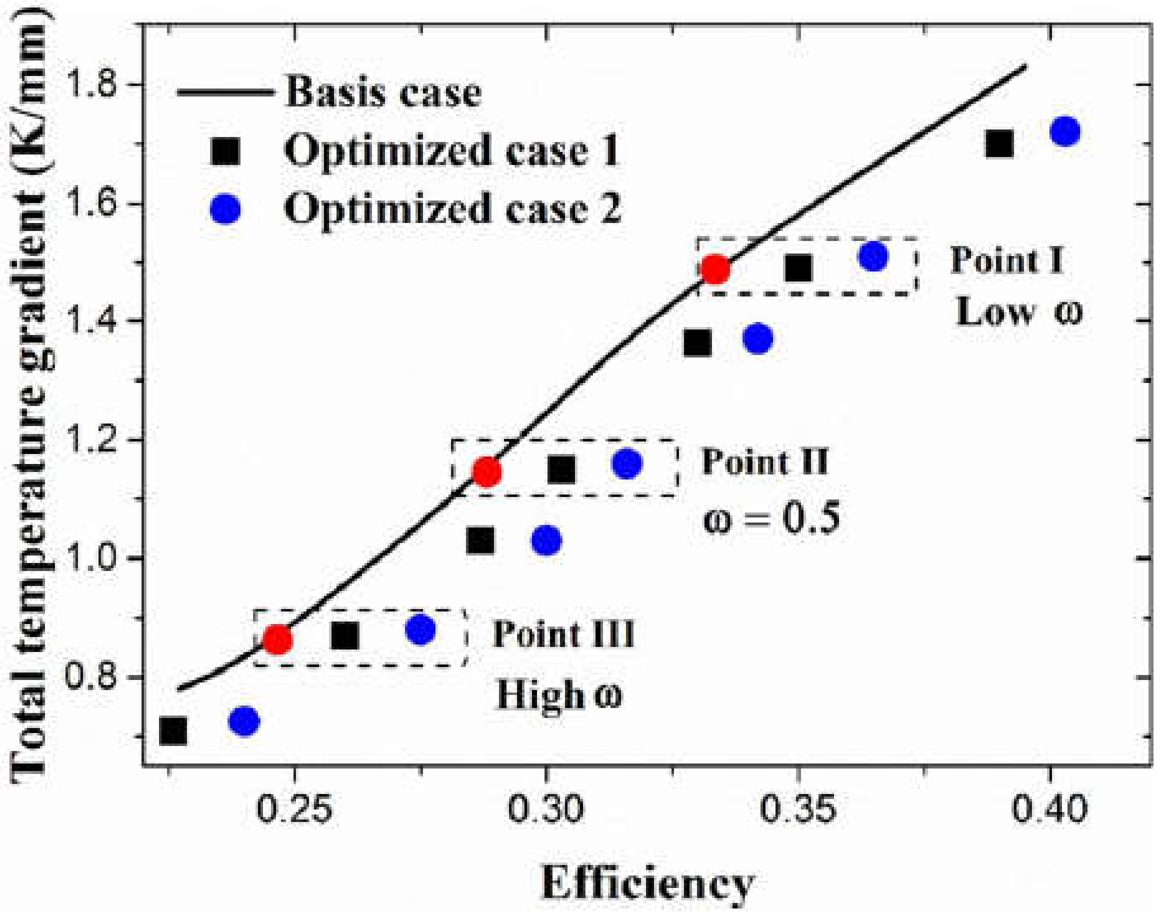
Table 3. Manipulating Variables and Their Ranges

operating variables		range/values
1	φ	10 to 20
2	ΔT_{in} (K)	70 to 120
3	y_{O_2}	0.21 and 0.25

Table 4. Optimization Designed with Different Schemes

optimization case (OC)	operating variables
OC 1	1, 2
OC 2	1, 2, 3

Optimisation results



Applications in SOFC Model Validation

Requirements of a validation scheme

- Quantification of measurement uncertainties
- Sensitivity analysis to identify parameters and inputs with significant influence on the measured output
- Experimental design to minimise the costly experimental runs
- Parameter estimation method to calibrate model parameters
- Statistical validation metric that accounts for model and measurement uncertainties

Why statistics based validation method?

- Necessary to account for uncertainties in model predictions (due to parameter uncertainties) and the uncertainties in measurements.

Basic concepts and assumptions

- The parameter uncertainty and the measurement uncertainty are normal distributions and not correlated.
- The probability density function for a normal distribution will be a bell curve. The same for a bivariate distribution will be a surface whose projections at different fixed probabilities will be ellipses. The general equation for a 2 dimensional probability distribution function is:

$$\mathrm{PDF}(a,b) = \frac{1}{2\pi\sqrt{|\mathbf{V}|}}\exp\left(-\begin{bmatrix} a - a_{mean} & b - b_{mean} \end{bmatrix}\mathbf{V}^{-1}\begin{bmatrix} a - a_{mean} \\ b - b_{mean} \end{bmatrix}\right)$$

Basic concepts and assumptions

- A constant r^2 is defined, which could be thought of as a distance measure (the radii of the ellipse).

$$r^2 = \mathrm{d}^{\mathrm{T}} \mathrm{V}^{-1} \mathrm{d}$$

where $\mathrm{d}^{\mathrm{T}} = \left[x_1 - x_1^{mean} \ldots x_n - x_n^{mean} \right]$

and **V** is the co-variance matrix defined as:

$$\mathrm{V} = \begin{bmatrix} \mathrm{cov}(a,a) & \mathrm{cov}(a,b) & \mathrm{cov}(a,c) \\ \mathrm{cov}(b,a) & \mathrm{cov}(b,b) & \mathrm{cov}(b,c) \\ \mathrm{cov}(c,a) & \mathrm{cov}(c,b) & \mathrm{cov}(c,c) \end{bmatrix}$$

$$\langle \mathrm{cov}(a,b) \rangle = \frac{1}{n-1} \sum_{i=1}^{n} \left(a_i - a_{mean} \right) \left(b_i - b_{mean} \right)$$

Methods for model validation

- Monte Carlo simulations based method
- Optimisation based method

Monte-Carlo based method

$$r^2 = \mathrm{d}^{\mathrm{T}} \mathrm{V}^{-1} \mathrm{d}$$

$$\mathrm{d}^{\mathrm{T}} = \left[M_1 - P_1^{mean} \quad \ldots M_r - P_r^{mean} \right]$$

$$\mathrm{V} = \left[\mathrm{V}_{\mathrm{prediction}} \right]$$

- Obtain samples from the parameter distributions and perform Monte-Carlo simulations.
- The means and covariance matrices for the prediction uncertainty (normally distributed) are obtained from these simulations and the measurement uncertainty.
- A critical value (r_{crit}^2) of the distance measure is defined from the chi square values (which is similar to the r^2). The chi square value corresponds to the sum of the squares of the individual normal distributions at the specified probability level.
- If r^2 obtained from above < r_{crit}^2, accept model as valid.
- Parameter estimation is independent of the validation process.

Drawback: Requires many thousands of costly simulations

Alternate: Optimisation based methods

Optimisation based validation method

$$r^2 = \mathrm{d}^{\mathrm{T}}\mathrm{V}^{-1}\mathrm{d}$$

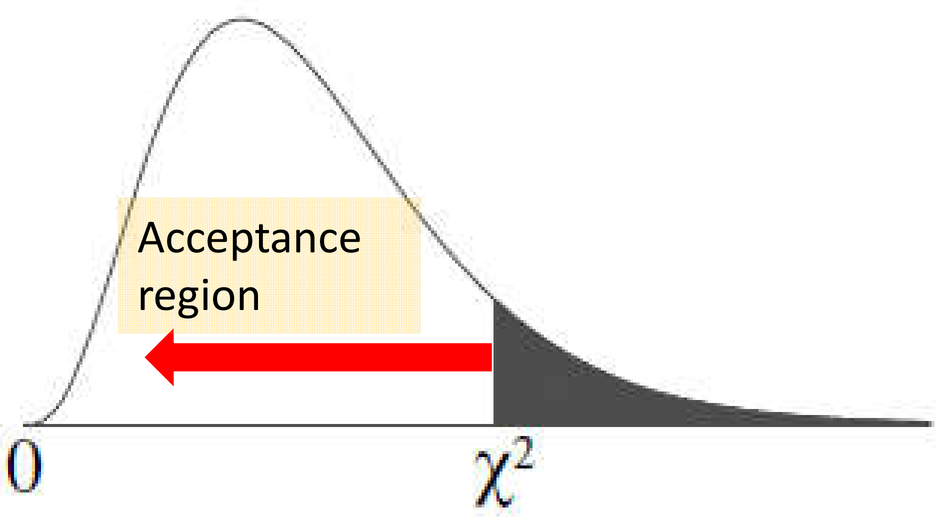
$$\mathrm{d}^{\mathrm{T}} = \left[\, p_1 - p_1^{mean} \quad \ldots p_n - p_n^{mean} \;\; P_1 - M_1^{mean} \quad \ldots P_r - M_r^{mean} \,\right]$$

$$\mathrm{V} = \begin{bmatrix} \mathrm{V}_{\mathrm{param}} & 0 \\ 0 & \mathrm{V}_{\mathrm{meas}} \end{bmatrix}$$

- The measurements are assumed to be a correct estimate of the measurement population means.
- A critical value of the distance measure for a particular confidence level is defined from the Chi square values.
- Optimisation is performed with the objective of minimising the distance measure.
- Accomplishes model calibration as well as validation.

If r^2 obtained from optimisation < r_{crit}^2, accept model as valid.

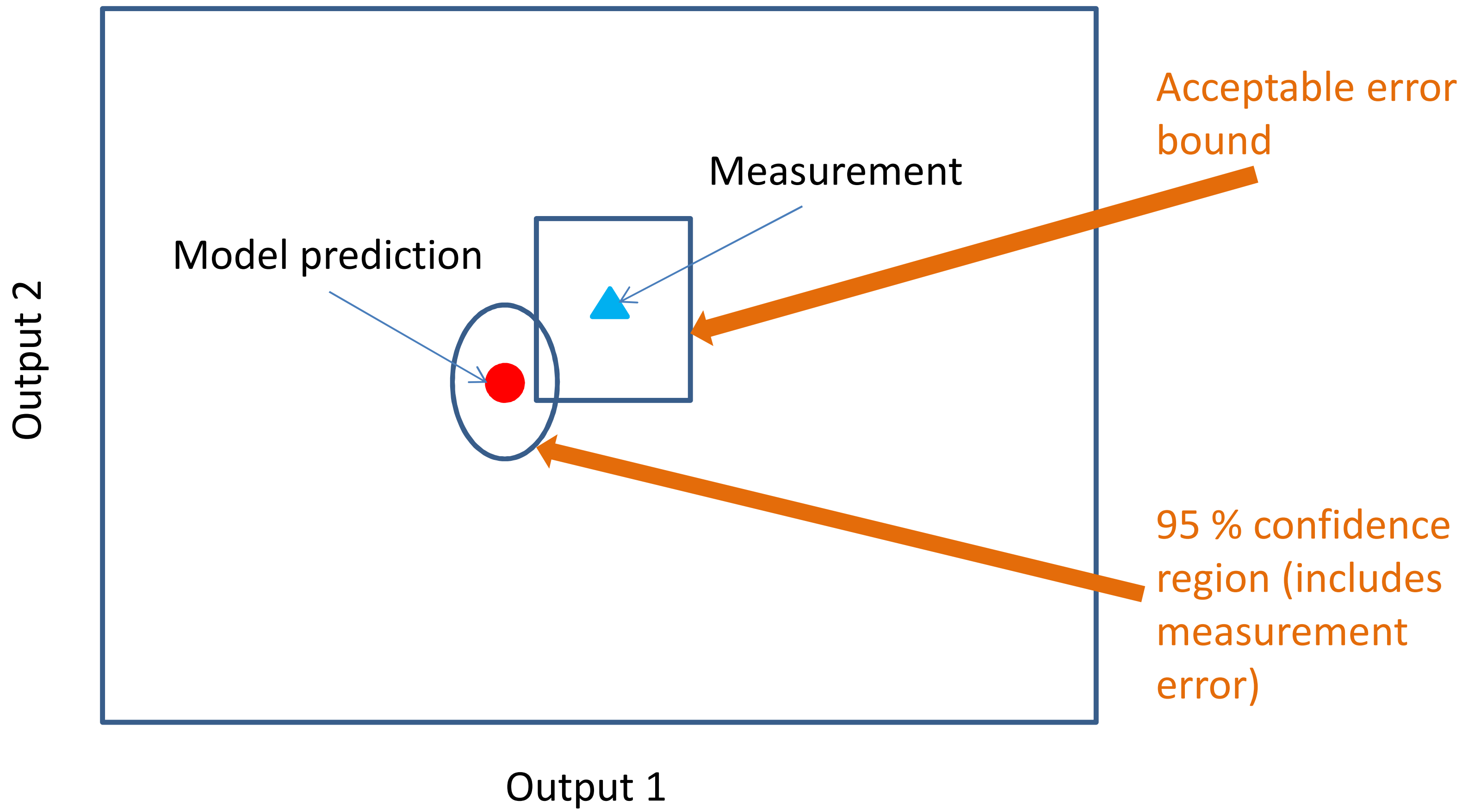
Statistical Hypothesis testing



α- Significance level (0.05) – probability of obtaining a sample as bad as or even worse than the current observation.

(1-α) (0.95) – Confidence level. More confidence in the result is obtained by having a lesser probability of erroneous sample.

Engineering validation



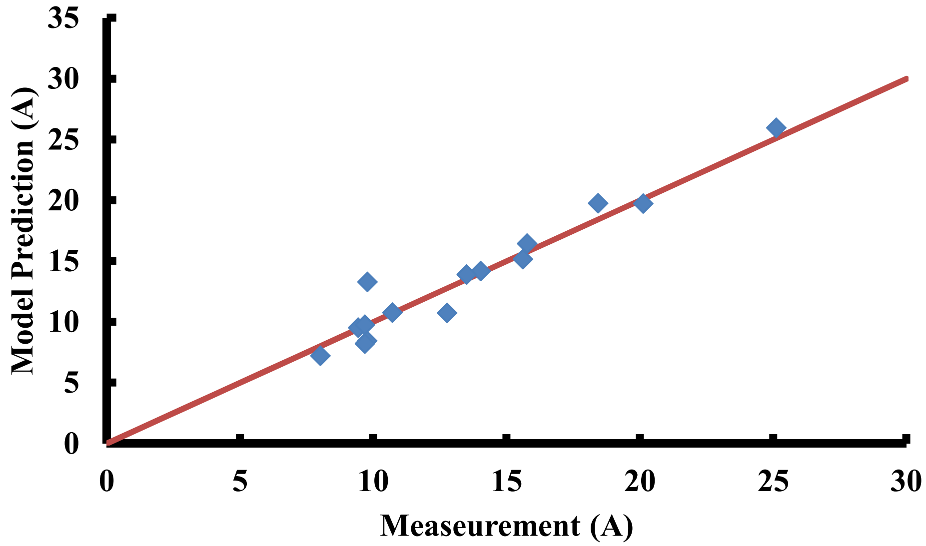
Two step engineering validation

Step 1: Minimise r^2 with model parameters as design variables.

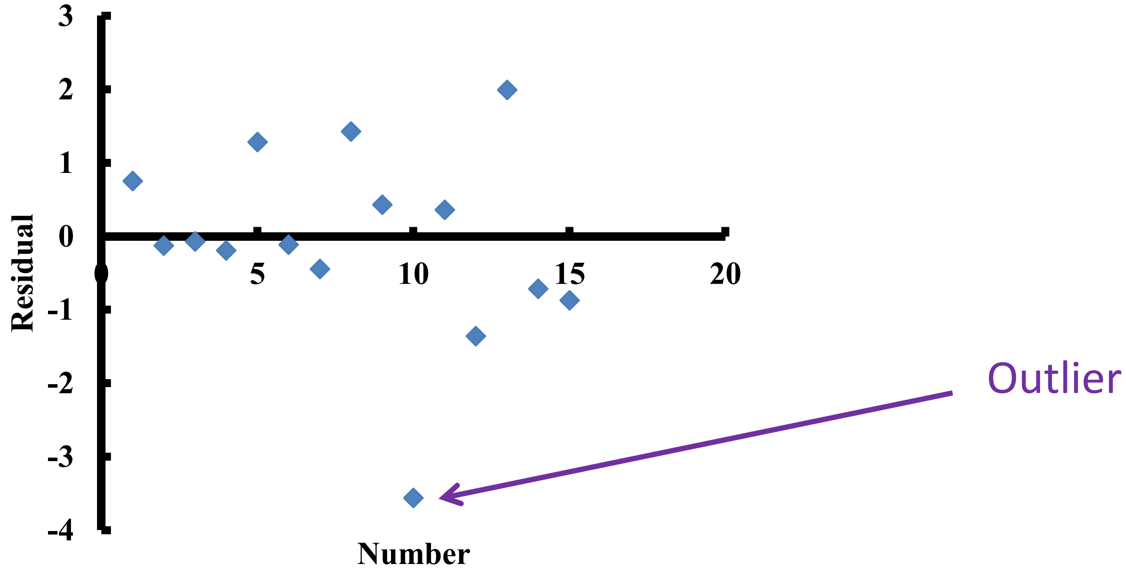
Step 2: Fix the model parameters at optimum values from step 1 and minimise r^2 with measurement values as design variables (with increasing acceptable error).

Graphical method for validation

Parity plot



Residual plot



Identified sensitive parameters

8	Cathode side, Pre-exponential kinetic factor (A/m^2)
10	Activation Energy, Cathode side, J/mol
25	H2O diffusion coefficient (1/s)

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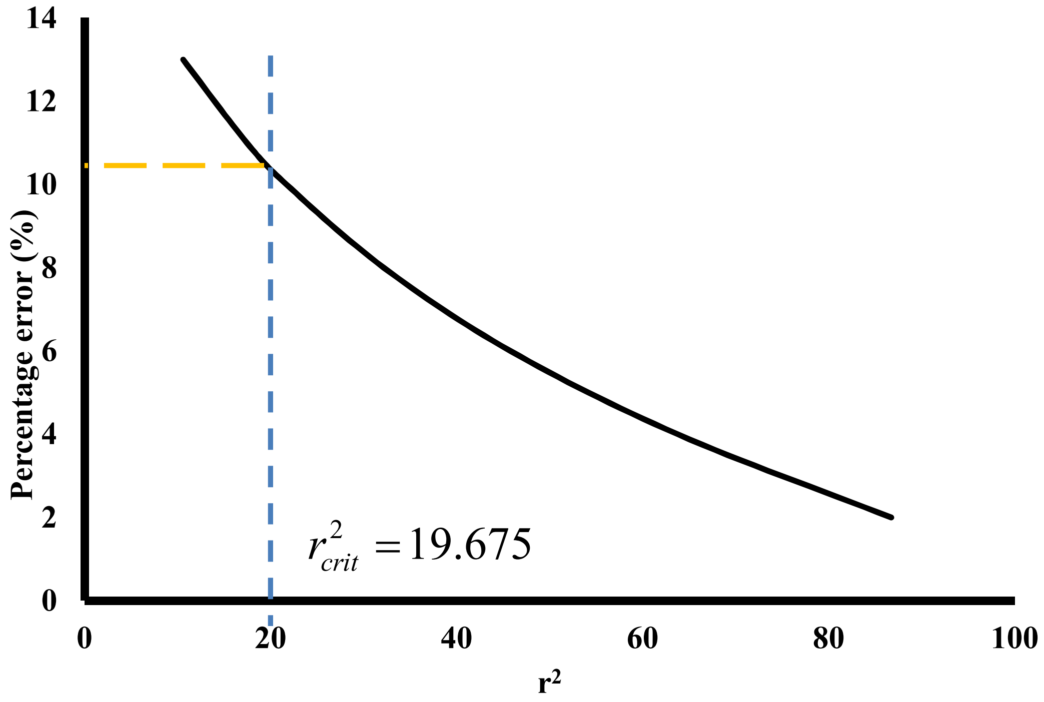
3 parameter result – Engg. Validation



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Sensitive parameters

1	Cathode side, Pre-exponential kinetic factor (A/m^2)
2	Activation Energy, Cathode side (J/mol)
3	Activation Energy, Anode side (J/mol)
4	Porosity
5	Anode side anodic transfer coefficient
6	Membrane conductivity pre-exponential coefficient (S/m)

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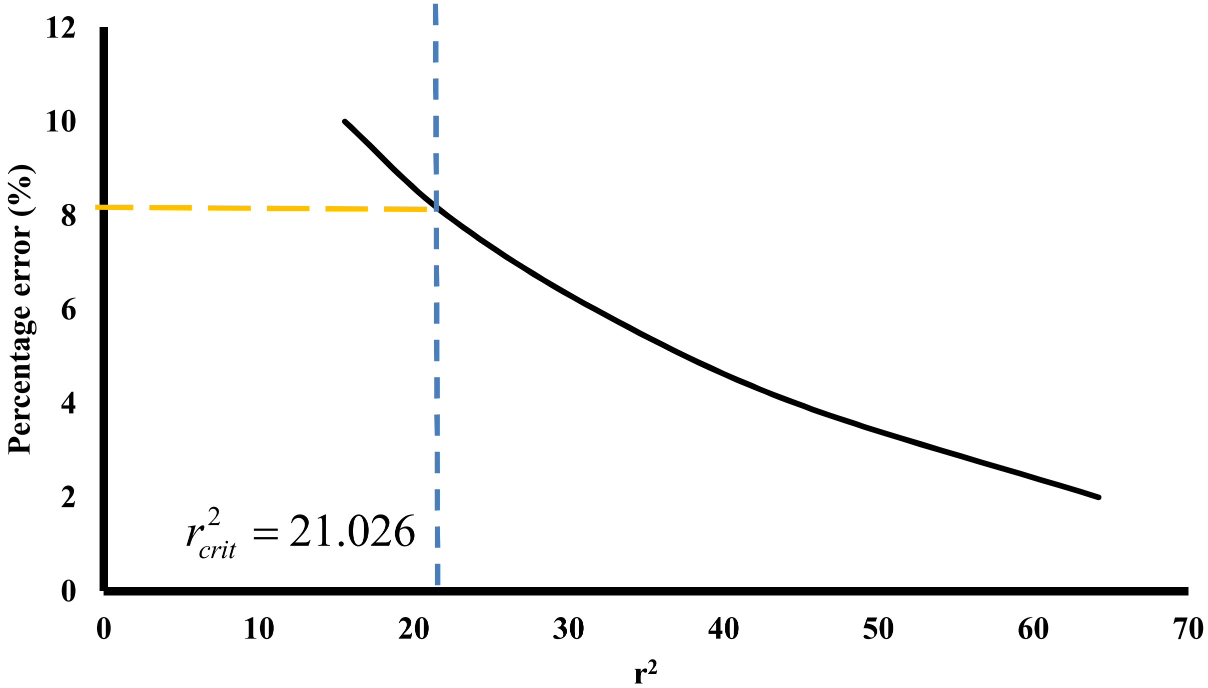
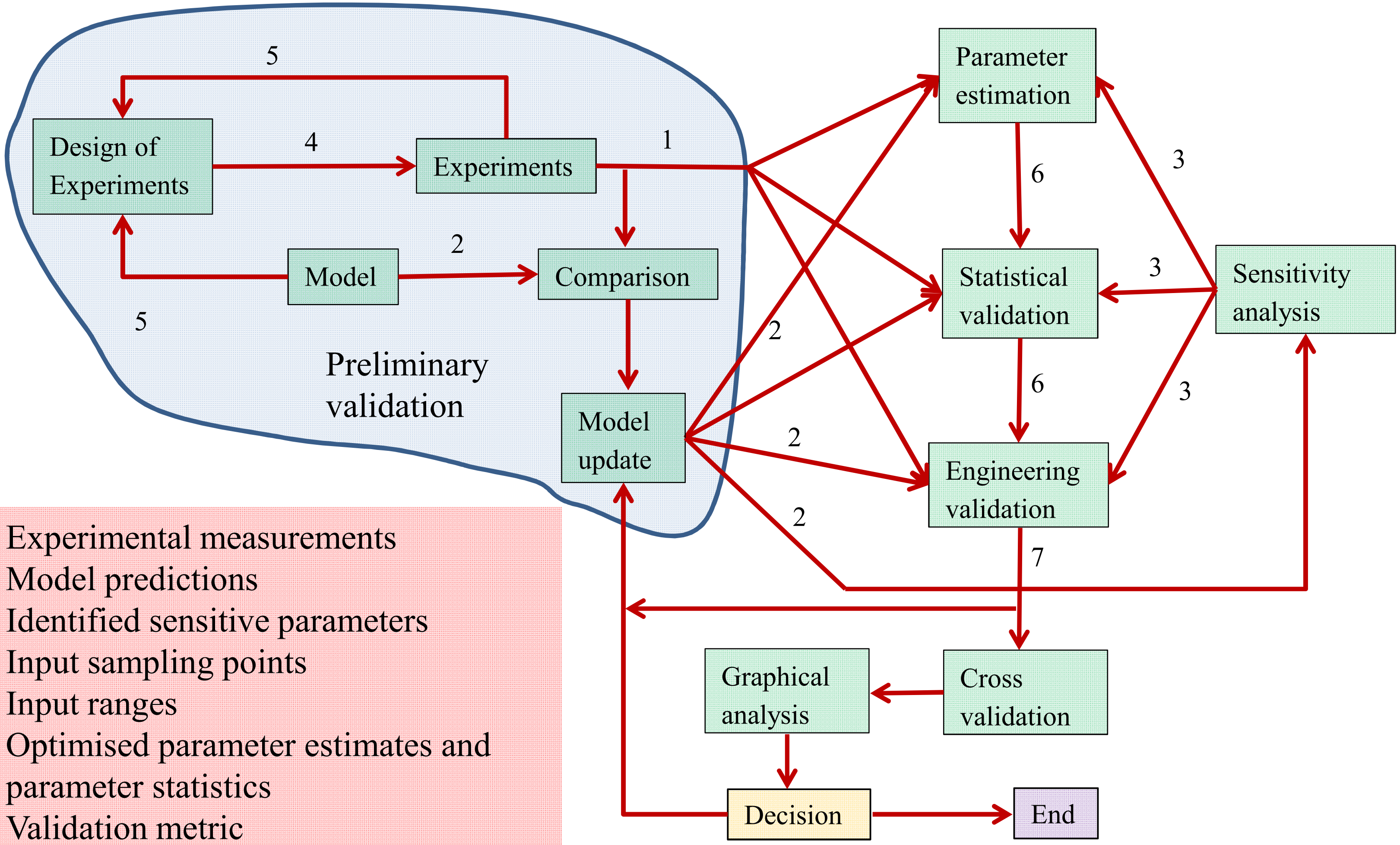
6 parameter result (cross validation)

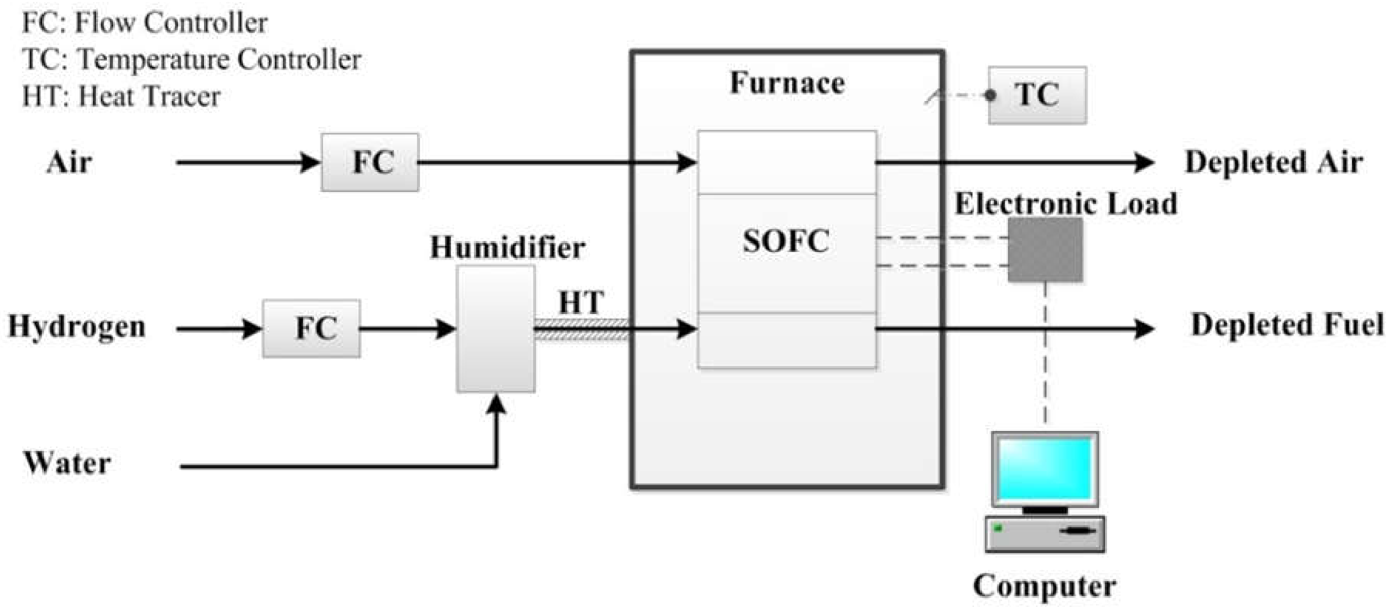


- A set of 6 data points used (different from original set of 8 points)
- Parameter estimates and mean retained from original case
- Model predictions correspond to the new set of data points

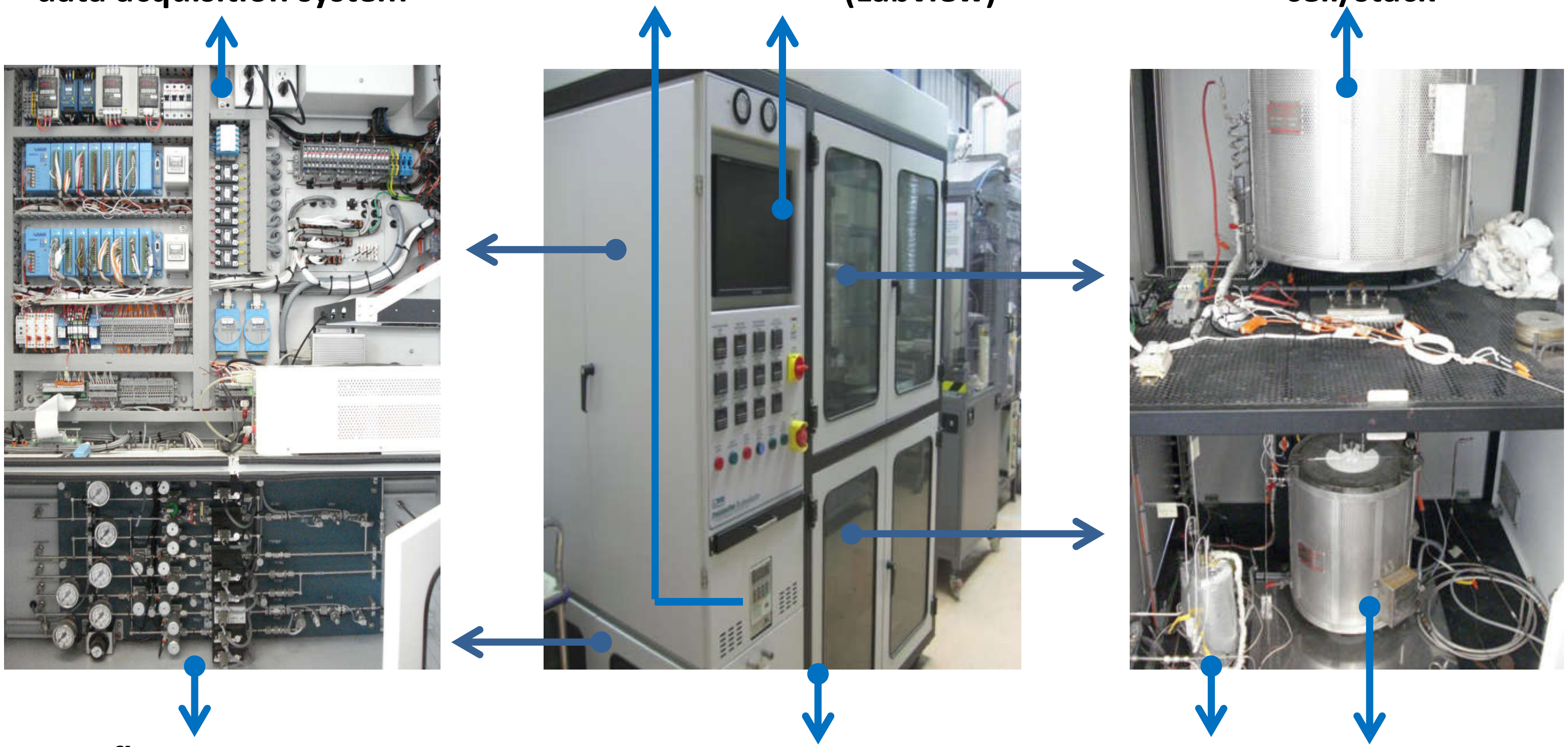


1. Experimental measurements
2. Model predictions
3. Identified sensitive parameters
4. Input sampling points
5. Input ranges
6. Optimised parameter estimates and parameter statistics
7. Validation metric

Schematic of the experimental setup



The SOFC test station at Curtin University



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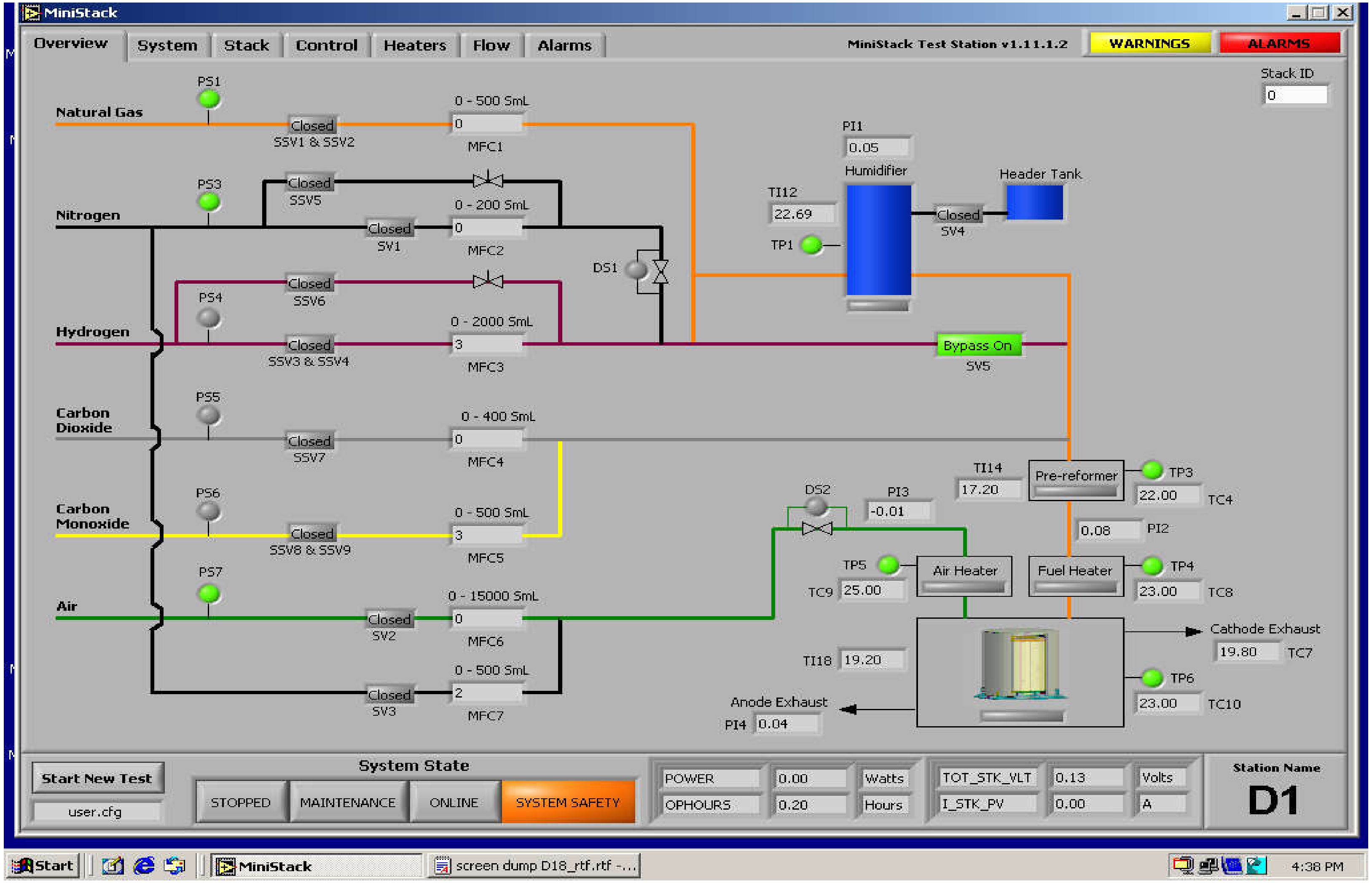
Schematic view for the monitoring system of the SOFC test rig

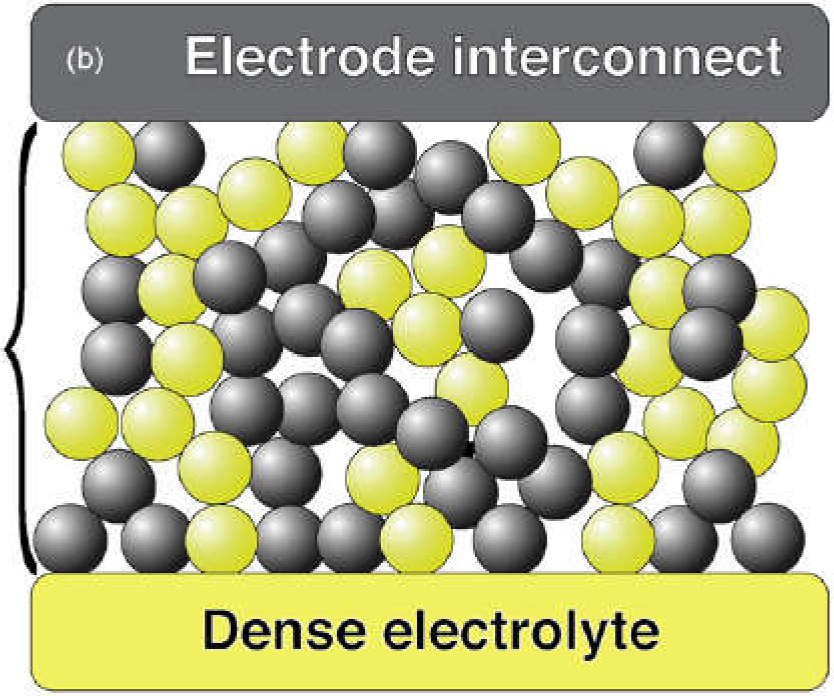


Features of the SOFC test station

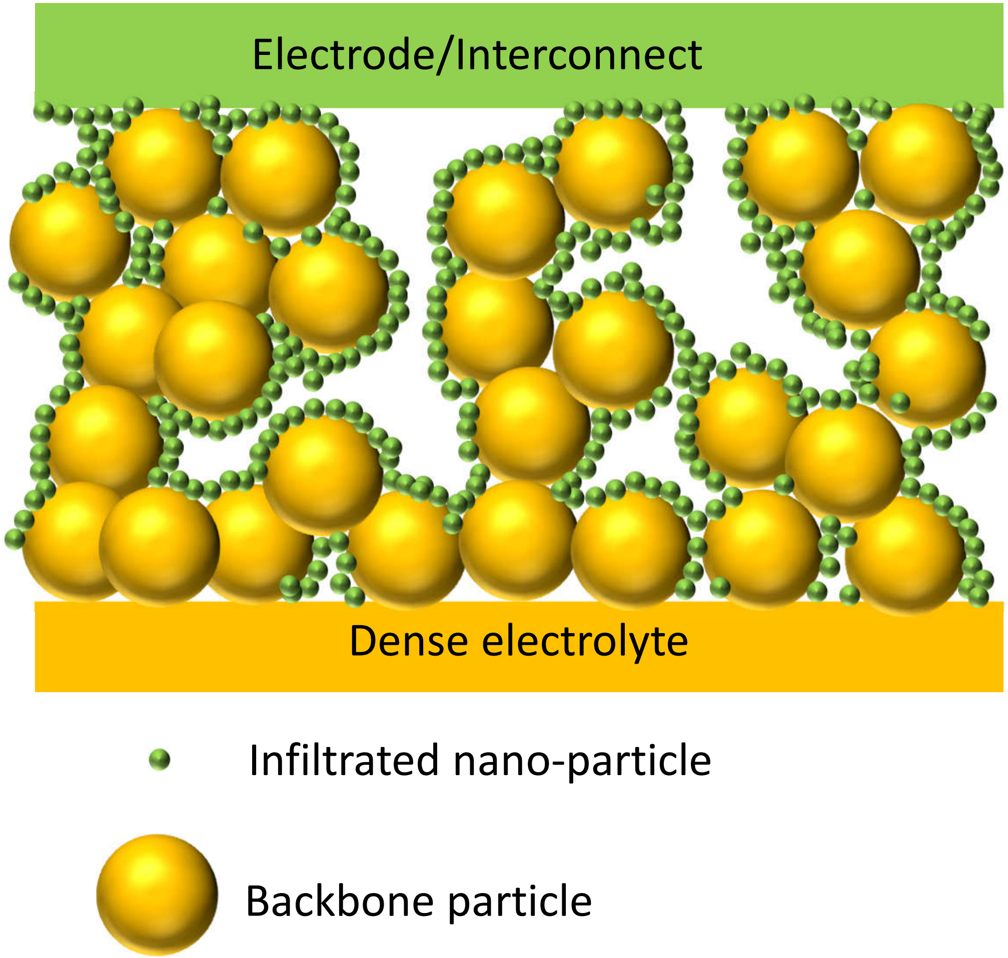
- The rig includes a furnace for placing the cell or stack, pre reformer, Humidifier, distilled water header tank, vent duct, a computer for human interface, programmable load, heaters, control valves and piping.
- Could be adopted to run either on pure hydrogen or methane. Can also include CO2, CO and N2 if required so that the cell could be run on a simulated gas mixture
- Contains 11 channels for voltage and temperature measurement.
- Allows both potentiostatic and galvanostatic operations of the fuel cell.
- Control and user interface based on Labview software.
- Safety features built into the system.

Electrode microstructure optimisation

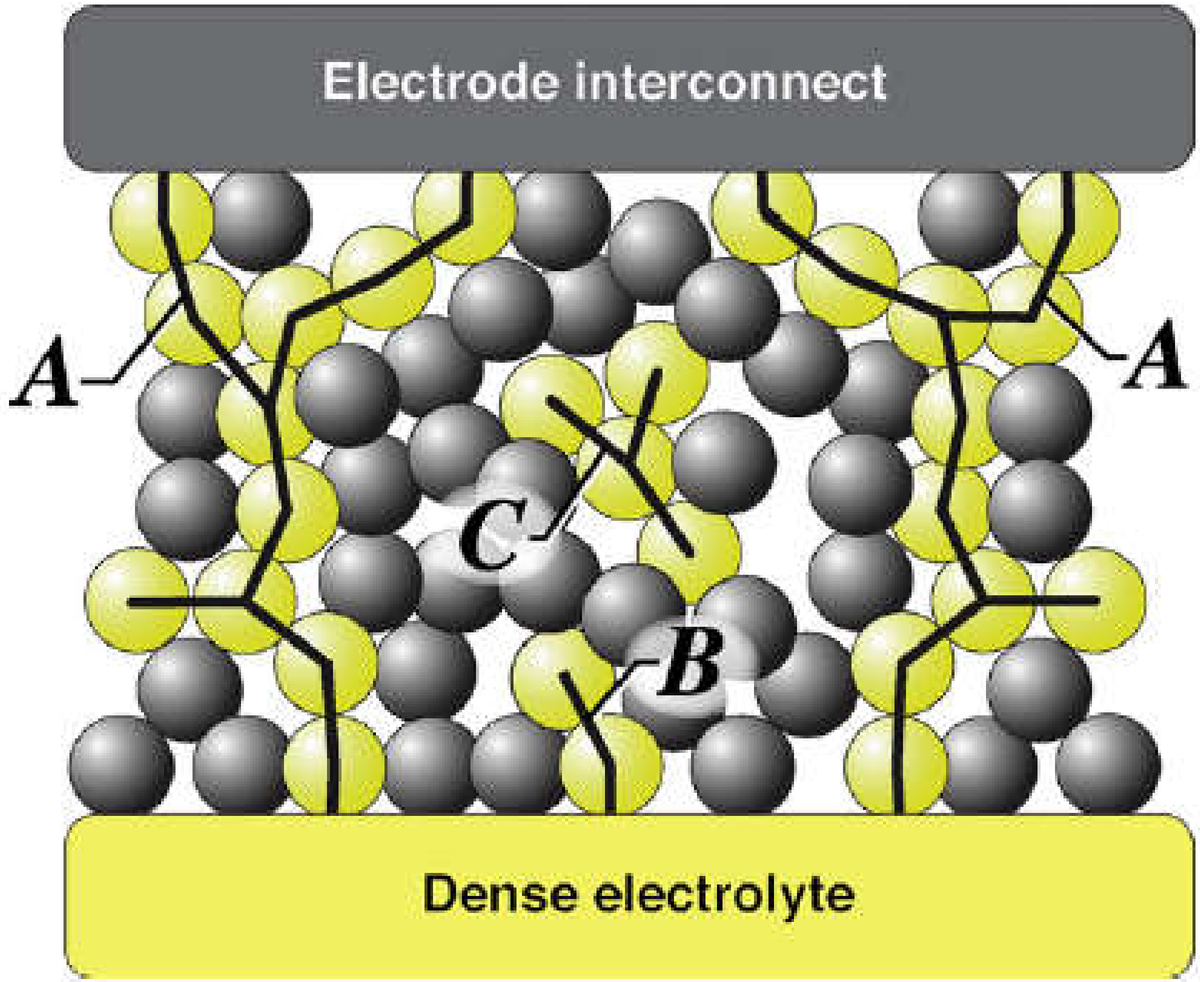
Typical microstructure with electrode and electrolyte particles



Electrodes infiltrated with nano-particles



Percolation paths in the electrode



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Microstructure affects performance

Microstructural Parameters
- Porosity
- Thickness
- Particle size ratio
- Size and Volume fraction of particles

→

Microstructural Properties
- Active surface area
- Average pore size
  Tortuosity
- Effective resistivity of ion and electronic conductor

→

Cell Performance

Polarisations (activation, concentration and Ohmic)

Electrode microstructure influence

Microstructural Parameters

- Porosity
- Electrode thickness
- Particle size ratio
- Volume fraction of particles
- Agglomeration effects

Microstructural Properties

- Triple Phase Boundary (TPB) area
- Active surface area
- Average pore size
- Tortuosity
- Effective resistivity of ion and electronic conductor

Effects of microstructure damage

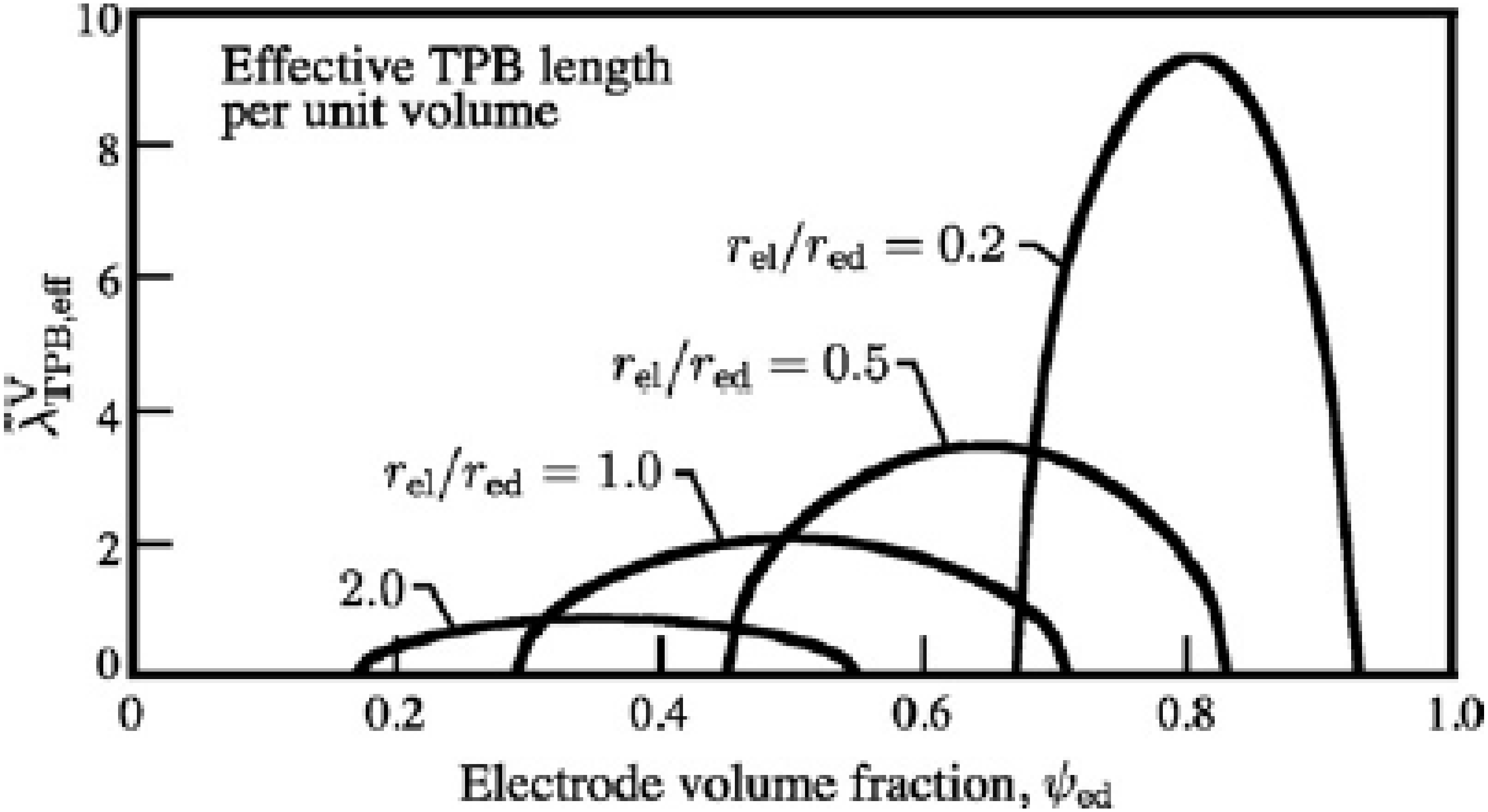
- Decrease in porosity
- Decrease in Active surface area
- Slower electrochemical reaction
- Electrode destruction

Electrode microstructure influence

Microstructure of anode ⟹ **Electrochemical performance**

Changes in the active sites for electro-chemical reaction	Activation polarization
Changes in the effective ionic and electronic resistivity	Ohmic polarization
Changes in electrode pore size	Concentration polarization

TPB length variation (traditional electrodes)



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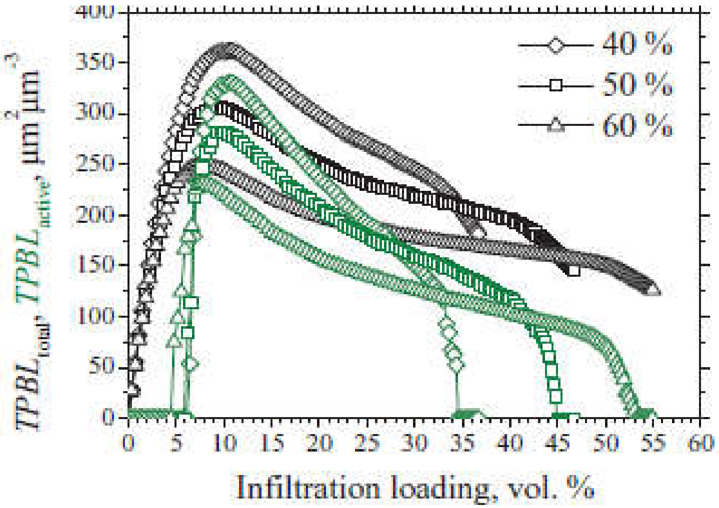
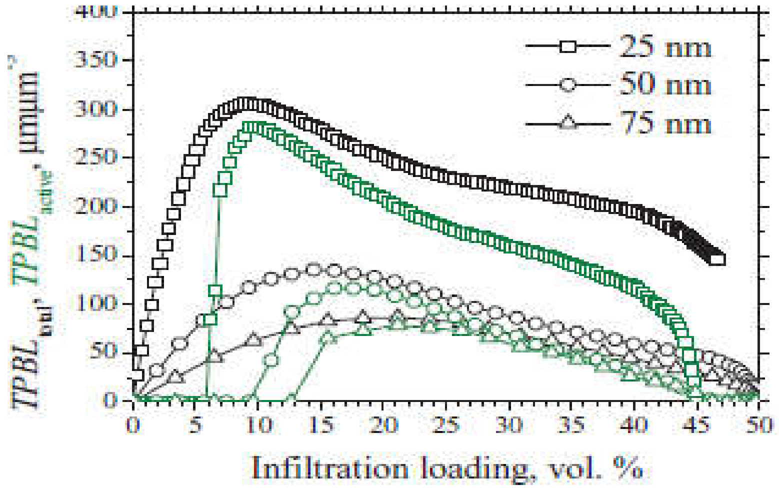
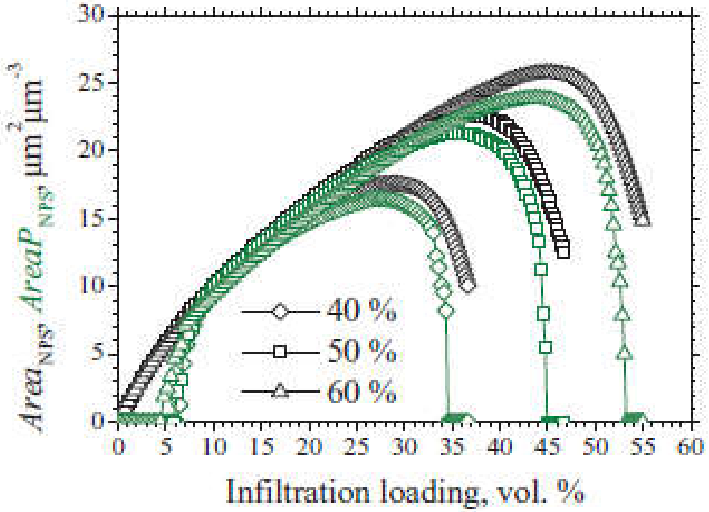
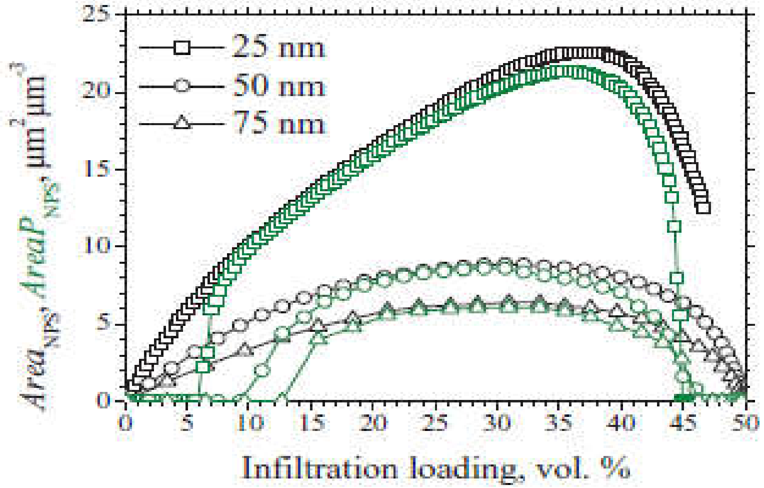
TPB length and surface areas(infiltrated electrodes)









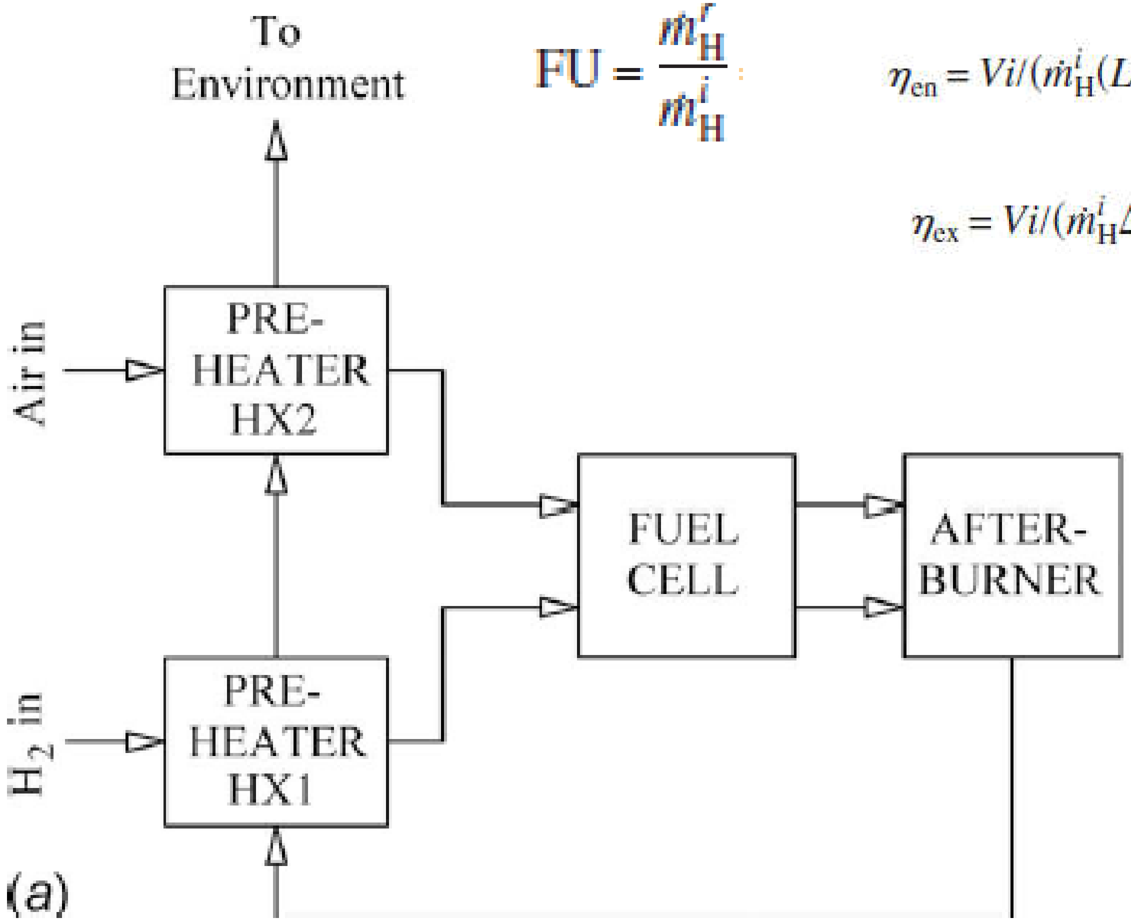
Energy and exergy optimisation

SOFC system with BOP components

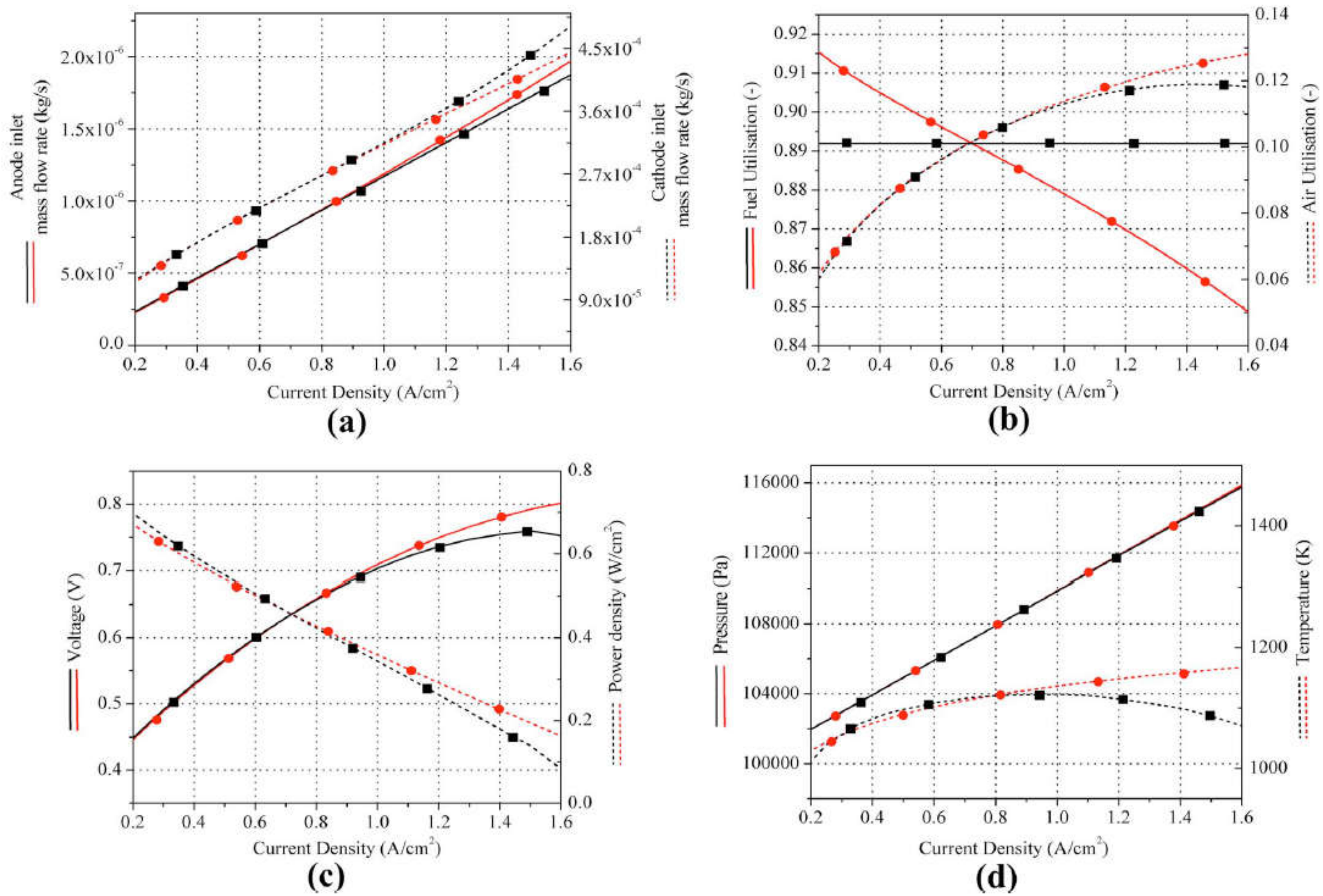
$$FU = \frac{\dot{m}^r_H}{\dot{m}^i_H}$$

$$\eta_{en} = Vi/(\dot{m}^i_H(LHV)_H)$$

$$\eta_{ex} = Vi/(\dot{m}^i_H \Delta G^0_T)$$



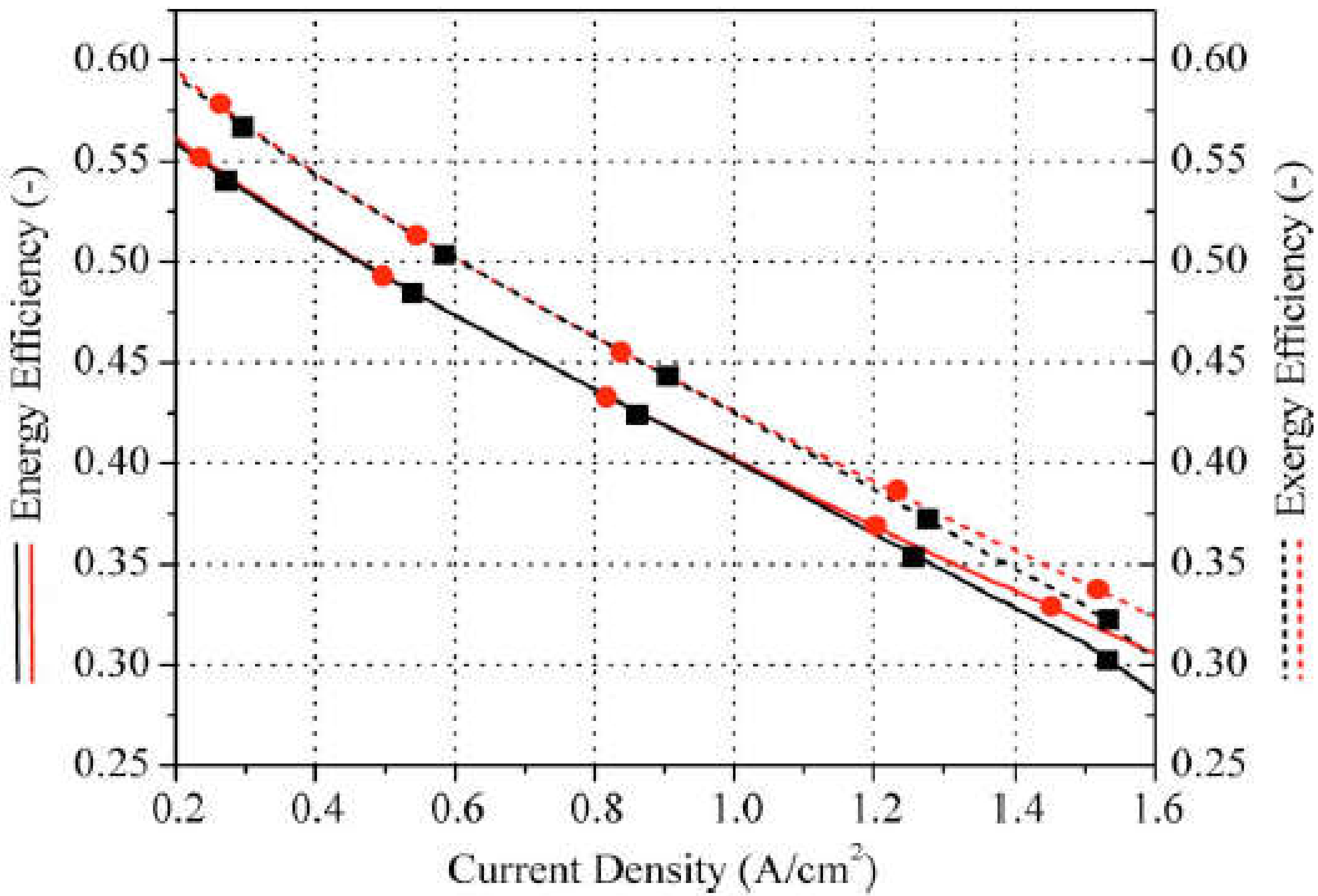
Operation under maximum efficiency and constant fuel utilisation modes



maximum efficiency (●) and constant FU (■) operations

Energy and exergy efficiencies

maximum efficiency (●) and constant FU (■) operations



constant FU operation of the fuel cell at a particular value of FU can closely approximate the maximum efficiency operation of the fuel cell

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Summary

- Optimisation methods have wide scope in improving the design and operation of SOFC systems
- Parametric study has limited scope as there is no guarantee of optimal solution
- Single objective optimisation is useful in some cases where there are no conflicting objectives
- Multi objective optimisation can provide trade-off optimal solutions considering conflicting design objectives and is very useful in the fuel cell field
- Quality of the model impacts the optimisation solutions and hence more effort is required for model validation and for improving model robustness

Some Relevant References

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YEARS 50 OF INNOVATION

Thank you

Make tomorrow better.