Metamodel-based optimization and parameter estimation for solid oxide cell stack development

A. Nakajo[a,b], F. Greco[a], T. Cornu[b], P. Caliandro[a], Z. Wuillemin[b], J. Van herle[a], C. Zeichmeister[c], S. Wolff[c], C. Bucher[c]

WOST Conference 2018

Weimar, Germany

21th June 2018

[a] Group of Energy Materials, Faculty of Engineering Sciences and Technology STI, Ecole Polytechnique Fédérale de Lausanne, Switzerland

[b] SOLIDpower-HTceramix, Yverdon-les-Bains, Switzerland

[c] Dynardo GmbH, Wien, Austria

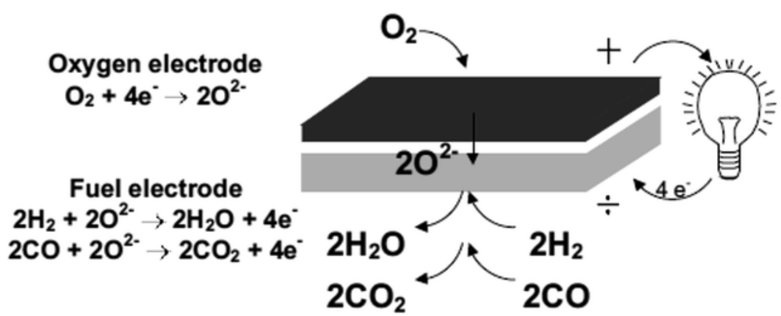
PROSOFC

Introduction

Solid Oxide Cell (SOC): direct energy conversion device (700-800°C)

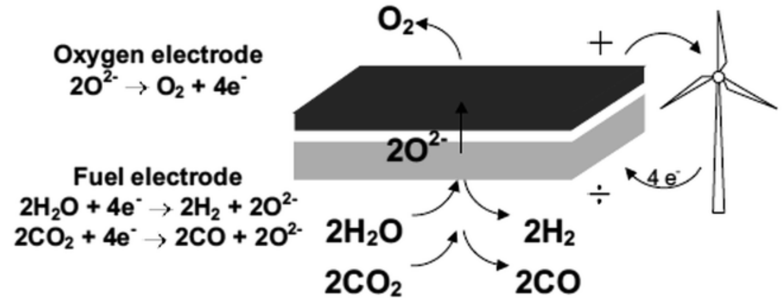
Solid Oxide Fuel Cell (SOFC):

- ✓ **75 % dc electrical efficiency** on natural gas.
- ✓ Long-term stability **< 0.33% / 1000 h**.



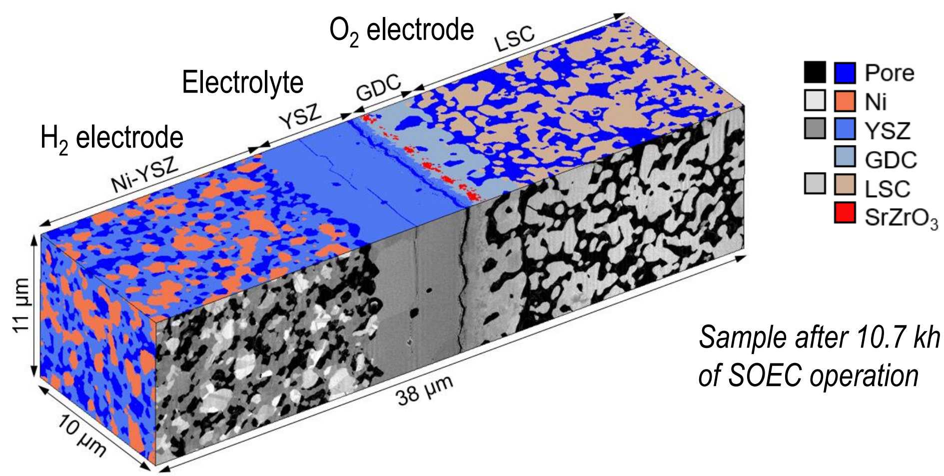
Solid Oxide Electrolysis Cell (SOEC):

- ✓ **Storing** growing **excess renewable electricity** into gas (H_2, CH_4).
- ✓ **95% conversion efficiency.**



- **Cell materials** (SOLIDpower)**:**

  YSZ: yttria-stabilized zirconia
  GDC: gadolinia-doped ceria
  LSCF: lanthanum strontium cobaltite ferrite

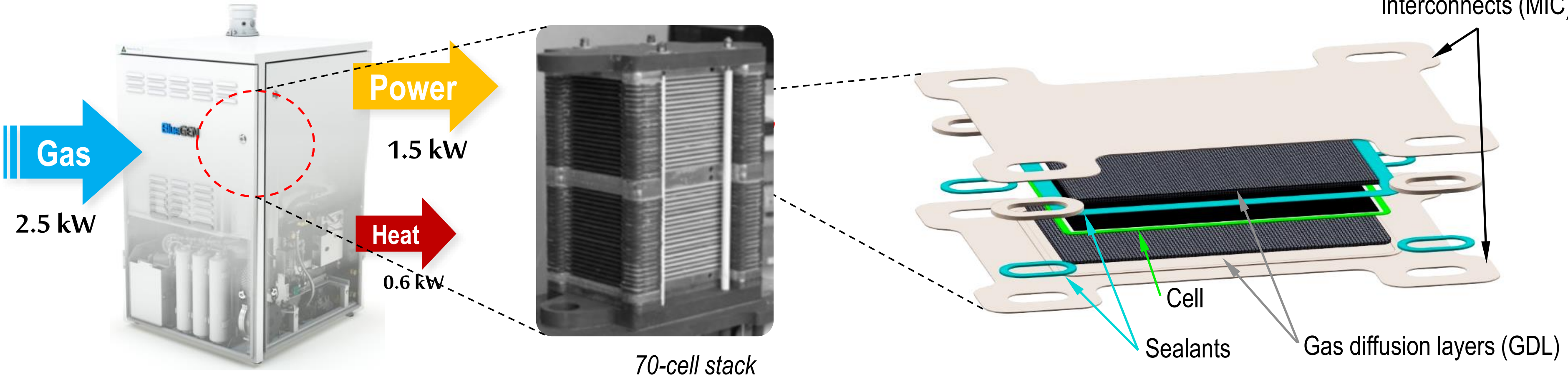


Sample after 10.7 kh of SOEC operation

PROSOFC

Introduction:

CHP fuel cell system | SOC stack | Repeating unit



70-cell stack

60% electrical efficiency
85% total efficiency

Example of applications (**SOFC mode**):

2.5 kW	5 kW	10+ kW
Residential	Schools Social Housing Buildings	Hotel Sport Center Data Center H_2 refueling stations Farms

Current challenge: mitigation of the remaining degradation:

- **Few % per year** (SOFC mode), increasingly difficult to detect, quantify and understand.

FP7 PROSOFC EU-PROJECT

PROSOFC

"Production and Reliability Oriented SOFC Cell and Stack Design"

M. Hauth et al., **Production and Reliability Oriented SOFC Cell and Stack Design,** *ECS Trans. 78 (1), 2231-2249 (2017).*

PROSOFC consortium:

1. **AVL GmbH** (Austria)
2. **HTceramix SA/SOLIDpower** (Switzerland/Italy)
3. **Dynardo GmbH** (Austria)
4. **Technical University of Denmark** (DTU)
5. **Forschungszentrum Jülich GmbH** (FZJ, Germany)

5. **Karlsruhe Institute of Technology** (KIT, Germany)
6. **Imperial College** (IC, Great-Britain)
7. **Joint Research Centre Petten** (JRC, Netherlands)
8. **EPFL** (Switzerland)
2. Topsoe Fuel Cell (Denmark)

Key addressed issues:

- Mechanical robustness and understanding of the interplay between material properties, stack design and operating conditions.
- Methodology for cost-optimal reliability-based design (COPRD) to guide the optimization of the cell and stack production (**Dynardo's software**).

EPFL SOLID POWER dynardo PROSOFC Schweizerische Eidgenossenschaft Confédération suisse Confederazione Svizzera Confederaziun svizra

Study cases

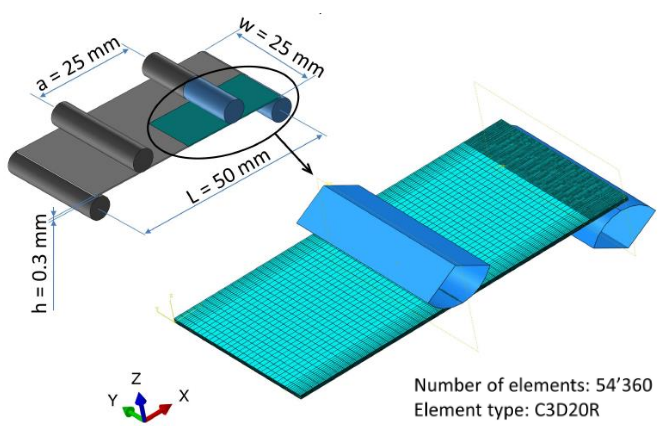
1. Measurement of the elastic, primary and secondary creep properties of cell materials by standard 4-point bending testing:
   - Metamodel-based parameter estimation for improved accuracy and flexibility in terms of constitutive laws, compared to processing by analytical solutions.

2. Metamodelling of the stack thermo-electrochemical behavior:
   - Optimization of the operation conditions for e.g load following (of the end-user demand).

Models interfaced with Dynardo's OSL/SoS

Finite-element model of 4-point bending (ABAQUS):

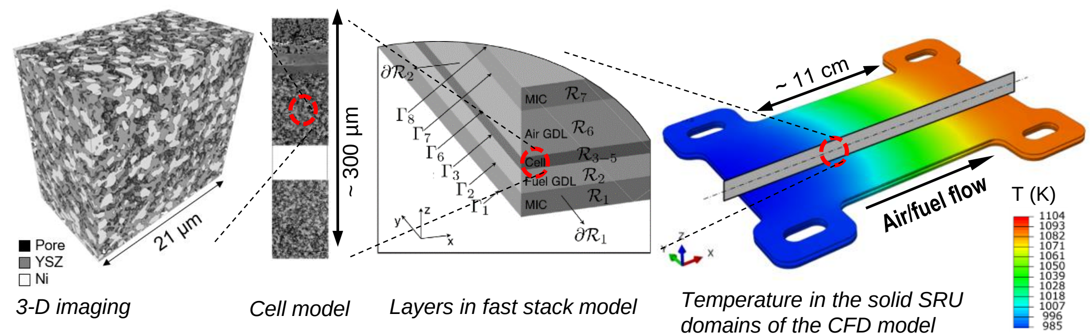
- **Testing intrinsic inaccuracy:**
  - Friction.
  - Anticlastic curvature.
  - Wedging stress.
  - Geometric non-linearity.
  - Contact point tangency shift.
- **Material non-linearity.**



Stack thermo-electrochemical model (gPROMS-FLUENT):

SOLIDpower stack:

- Close to 1-D temperature and overpotential profiles along the flow path.
- Model combination for fast simulations.



gPROMS — FLUENT

Local 1-D electrochemical model:

- Continuum electrode models.
- Distributed charge transport and transfer.
- Effective material properties measured by 3-D imaging.

Fast stack model:

- 1-D transport along fow path.

3-D CFD model:

- Sink/source terms from fast stack model.
- Periodic boundary conditions / y-symmetry.
- Discretized surrounding insulation and gas distribution domains.

EPFL SOLID POWER dynardo PROSOFC

Parameter estimation

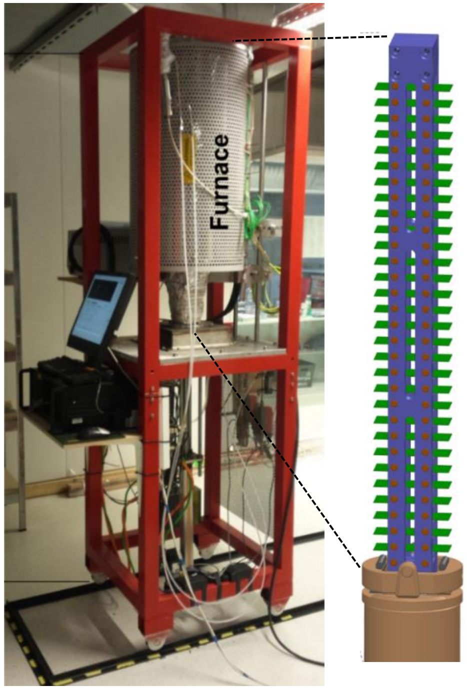
High-temperature setups for standard 4-point bending testing:

Elastic properties and strength:

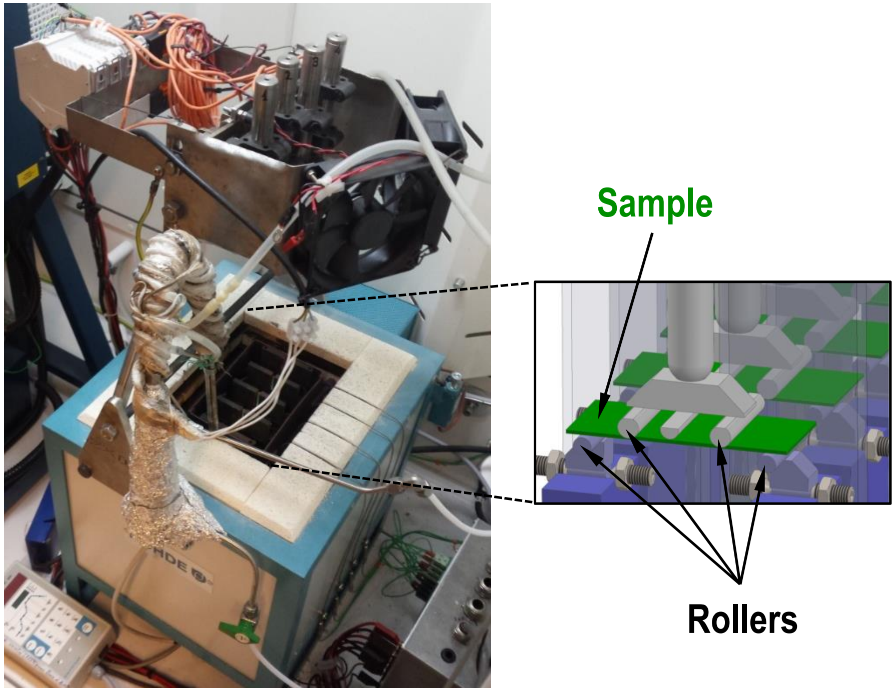
- Testing of up to 30 samples per heating cycles.
- Reducing, oxidising and humid atmospheres.

Creep properties:

- Simultaneous testing of 4 samples.
- Reducing, oxidising and humid atmospheres.



Similar equipment at DTU



➢ Focus on the **Ni(O)-YSZ** H_2 electrode material.

Parameter estimation workflow

Sensitivity analysis:

1. Design of experiments.

2. FE model simulations.

3. Retrieval of the responses.

Elastic properties:
a. Force vs. displacement
Responses:
- Loading.
- Unloading.
- **Δ(Loading - Unloading).**

Creep properties:
b. Creep deformation vs. time
Responses:
- Creep deformation at different loads.

Model runtime: ~ hours

F-MOP generation:

$$RF = \mu + \sum_{i=1}^{n} z_i \, \phi_i$$

Mean

Array of the amplitudes relative to $\emptyset_i$

Shape function *i*

a. **Components of RF (random-fields)** computed from the sensitivity anaylsis signals.

b. **F-MOP** relative to $\emptyset_i$ **generated from the amplitudes** Z_i.

Model runtime: ~ seconds

Optimization:

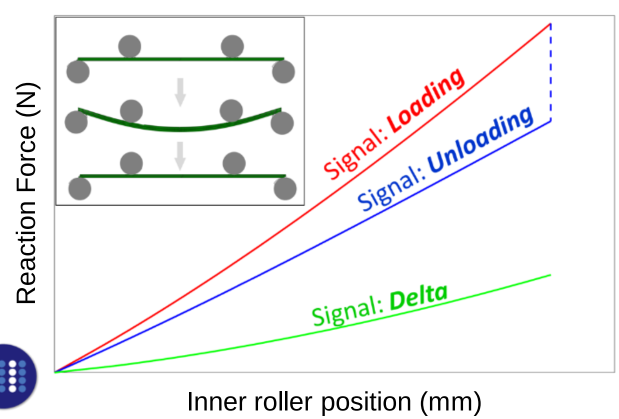
a. Objective function: difference **between the simulated RF** and the **experimental data**.

b. Optimisation using the **F-MOP**.

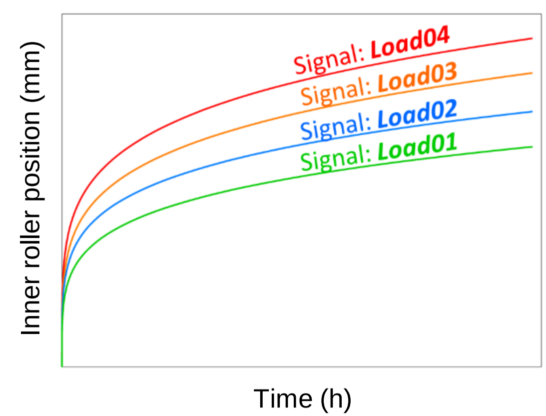
➔ **Parameter estimation with distributed metamodels**

Model runtime: ~ minutes

a. Elastic properties and **coeff. of friction**:



b. Primary and **secondary creep** properties:



EPFL SOLID POWER dynardo PROSOFC

Verifications

- **Numerical experiments:**
  - Variations of **target parameters, DoE sampling** and **optimization starting points:**

Elastic modulus and coefficient of friction:

Test	Start point of optimisation			Set of parameters to estimate	
	Em [GPa]	μ [-]	hs [μm]	Em [GPa]	μ [-]
#1	39.0	0.05	280	**100.0**	**0.7**
#2	209.0				
#3	39.0	0.99			
#4	209.0				

Creep parameters:

Test	Start point of optimisation						Set of parameters to estimate		
	A [$h^{-1}MPa^{-n}$]	n [-]	m [-]	Em [GPa]	μ [-]	hs [μm]	A [$h^{-1}MPa^{-n}$]	n [-]	m [-]
#1	1.28E-07	0.55	-0.05	100	0.7	280	**8.00E-07**	**1.5**	**-0.3**
#2	4.80E-06	2.4							
#3	1.28E-07	0.55	-0.78						
#4	4.80E-06	2.4							
#5	1.28E-07	0.55	-0.05	200.0	0.1	220	**2.00E-06**	**2.0**	**-0.7**
#6	4.80E-06	2.4							
#7	1.28E-07	0.55	-0.78						
#8	4.80E-06	2.4							

✓ **Input parameters** retrieved in all the tests with < 3% error.

- **Comparison with computational homogenization (elastic properties):**
  - Computational domain: 3-D electron microscopy.
  - Boundary value problem solved at the micro-scale with kinematic uniform boundary conditions.

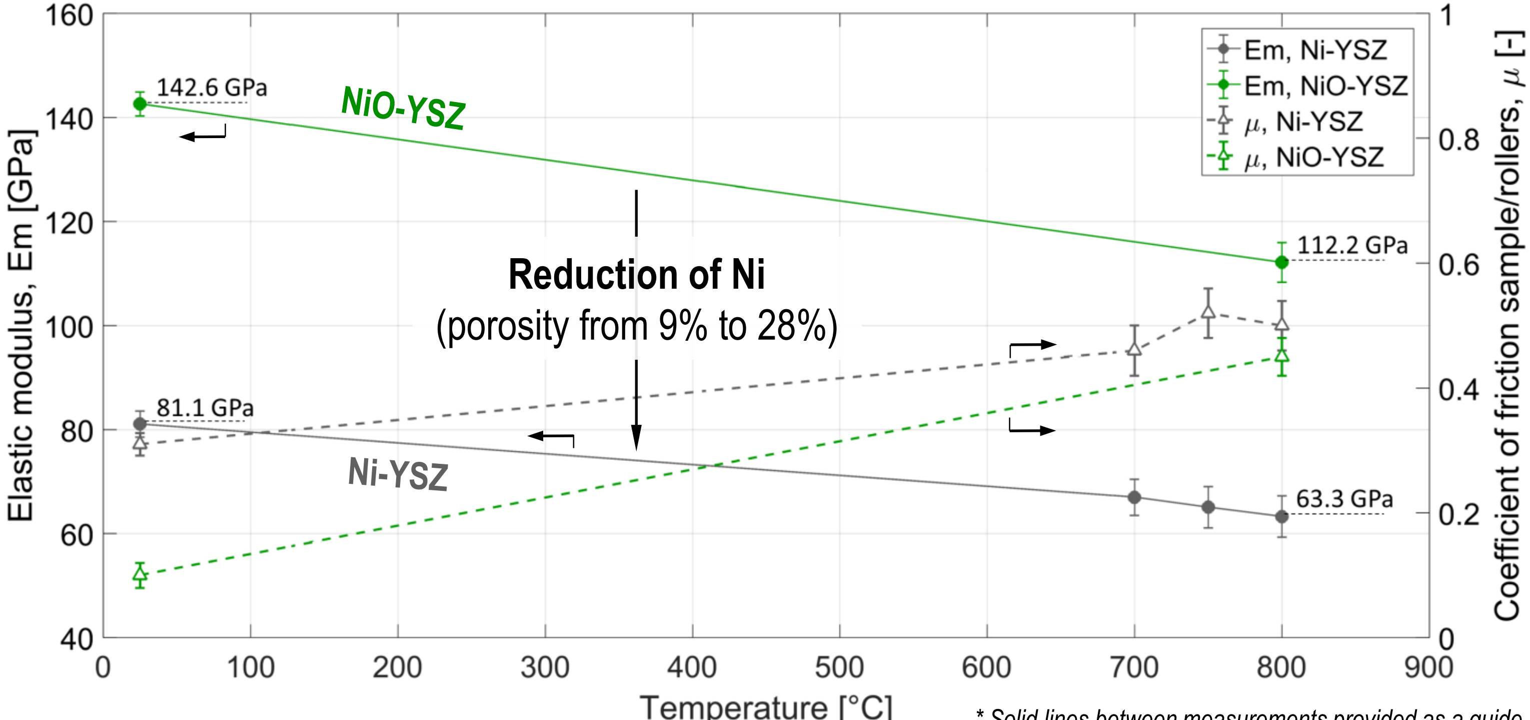
➢ Approximately **within the uncertainty** on the properties of the **YSZ** and **Ni** phases.

	E (GPa) 25°C	E (GPa) 800°C
4-point bending	81±2	63±3
Computational homogenization*	86±1	69±2

** Standard deviation: 4 x 9^3 μm^3 FIB-SEM volume samples.*

EPFL SOLID POWER dynardo PROSOFC

Ni(O)-YSZ elastic properties



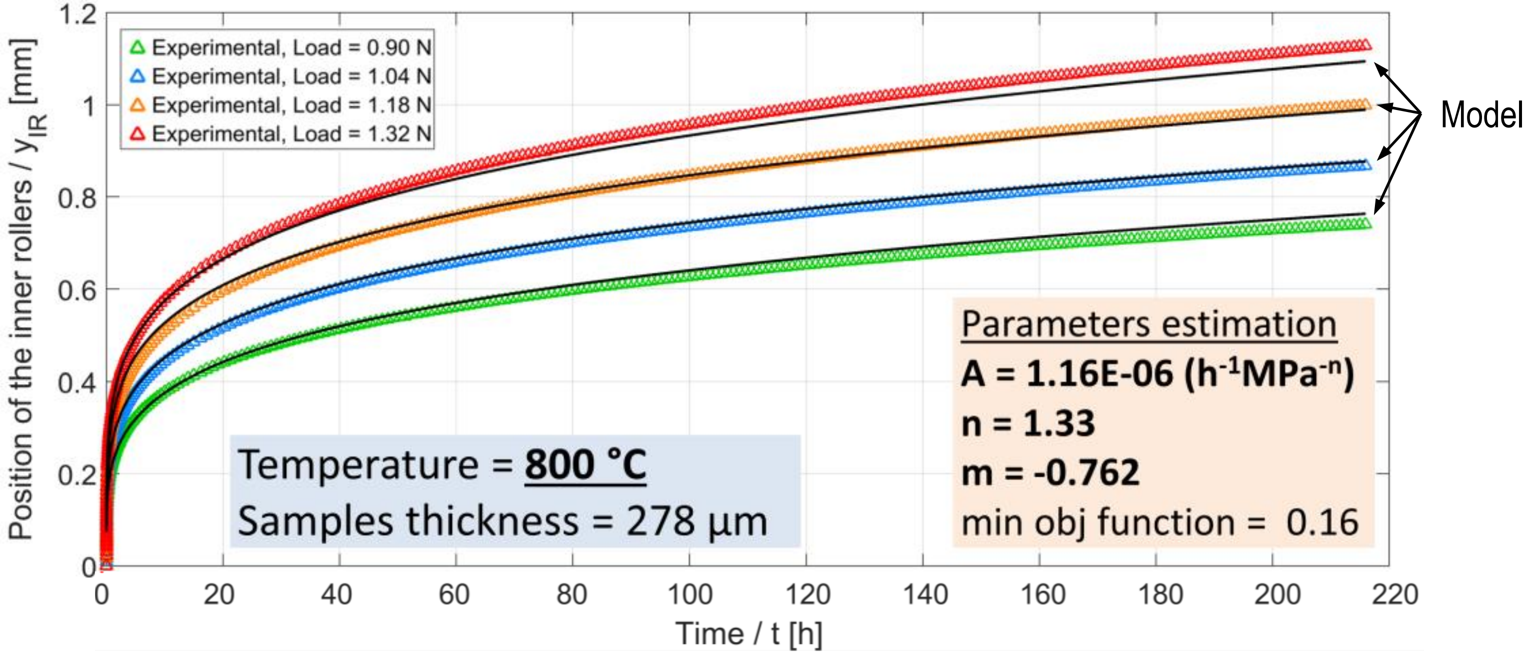
** Solid lines between measurements provided as a guide, not representative of the full temperature dependence.*

- Temperature dependence of the coefficient of friction, higher for Ni-YSZ:
    - Drying of the grease (contact interface) and potentially testing sequence.
- Processing by analytical solution: limited overestimation by ~10% (compensating effects).
- Accuracy not sufficient for aging analysis of the SOLIDpower Ni-YSZ (4700 h).

PROSOFC

Primary and secondary creep of Ni-YSZ

- **Measurements-model comparison:**



Strain-hardening creep model:
$$\dot{\varepsilon}_{ij}^{crp} = \frac{3}{2} \sqrt[m+1]{A\left[(m+1)\varepsilon_{eq}^{crp}\right]^{m} \left(\sigma_{eq}\right)^{n-1-m}} s_{ij}$$

Temperature dependence (700, 750, 800 °C):

$$A = Be^{-Q/RT}$$

B = 2.75 $MPa^{-n}\ h^{-1}$	m = [-0.76 ; -0.72]
Q = 130 ± 14 kJ mol^{-1}	n = [1.04 ; 1.33]

PROSOFC

Study cases

1. Measurement of the elastic, primary and secondary creep properties of cell materials by standard 4-point bending testing:
   - Metamodel-based parameter estimation for improved accuracy and flexibility in terms of constitutive laws, compared to processing by analytical solutions.

2. Metamodelling of the stack thermo-electrochemical behavior:
   - Optimization of the operation conditions for e.g load following (of the end-user demand).

Thermo-electrochemical metamodelling

Scalar metamodels, CoP matrix:

	Fuel utilization (-)	Max. T (K)	Current density (A cm^{-2})	Air inlet temperature (K)	Fraction of pre-reforming (-)	Steam to carbon ratio (-)	Interconnect thickness (m)	Aspect ratio of active area (-)	Total
Cell voltage (V)	6.4 %	3.9 %	82.1 %	1.0 %	0.6 %		2.0 %	0.7 %	95.5 %
Temperature difference (K)		11.7 %	31.9 %	21.3 %	1.1 %		34.6 %	11.4 %	98.4 %
Power density (W cm^{-3})	3.0 %	1.5 %	36.6 %				57.4 %		96.7 %
Electical efficiency (-)	9.9 %	6.2 %	78.3 %				4.0 %	1.1 %	95.5 %
Indicators for the risk of **Ni reoxidation** (fuel starvation)	40.2 %	0.0 %	52.2 %	0.0 %	0.0 %	2.1 %	0.0 %	0.8 %	96.0 %
	31.5 %	0.0 %	51.6 %	0.0 %	0.0 %	2.3 %	0.3 %	1.4 %	92.8 %
	37.1 %	0.0 %	51.8 %	0.0 %	0.0 %	1.9 %	0.1 %	0.6 %	96.4 %

Operation conditions:
~ Inlet gas flow, temperature and composition.
~ Polarization.

Simple design parameters

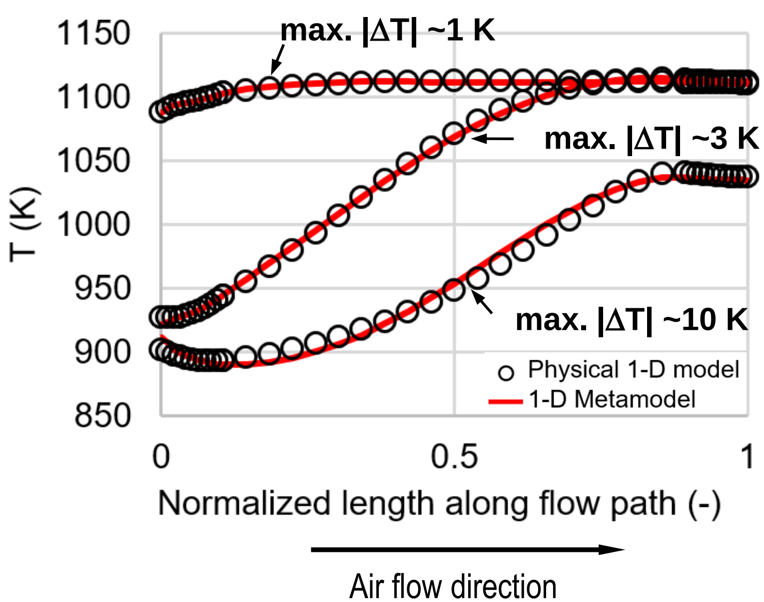
- **DoE with 150 samples (132 successfull).**
- **Large operation window.**
- **Parameter definition for accurate results.**
- **Initialisation sequence for low simulation failure rate.**

Thermo-electrochemical metamodelling

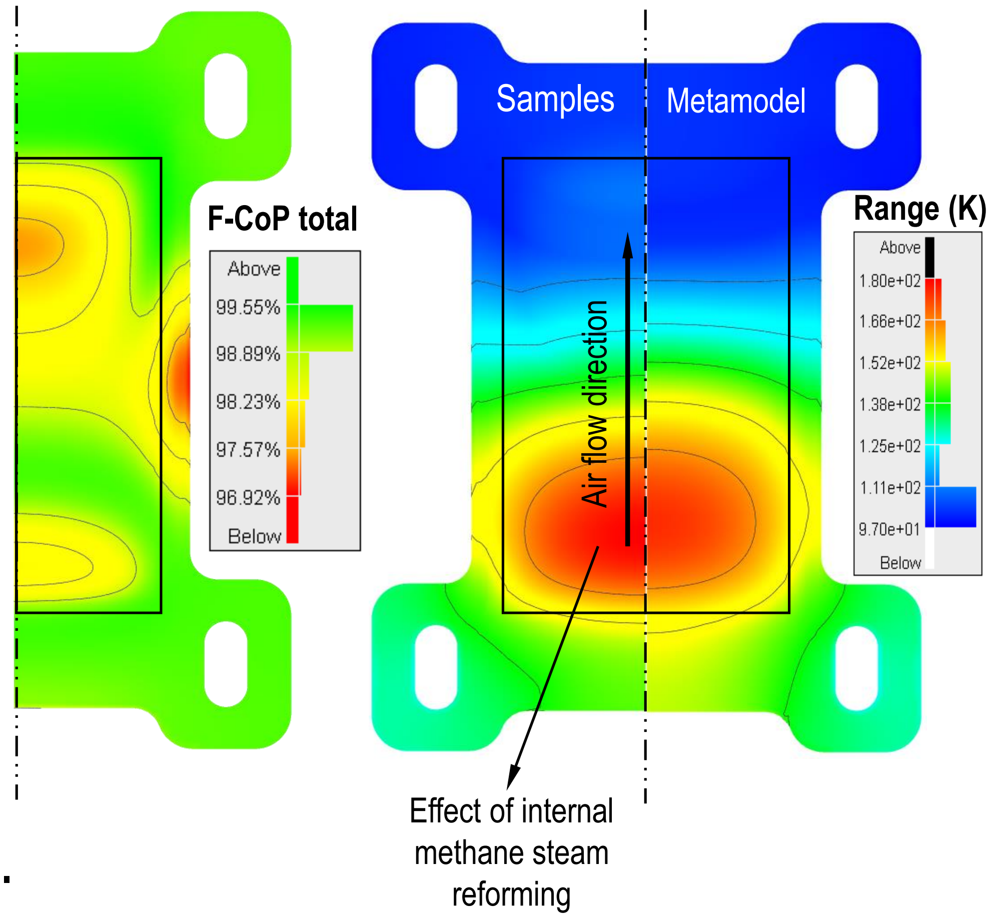
Temperature profile:

1-D stack model vs. metamodel:

Eample: 3/150 conditions



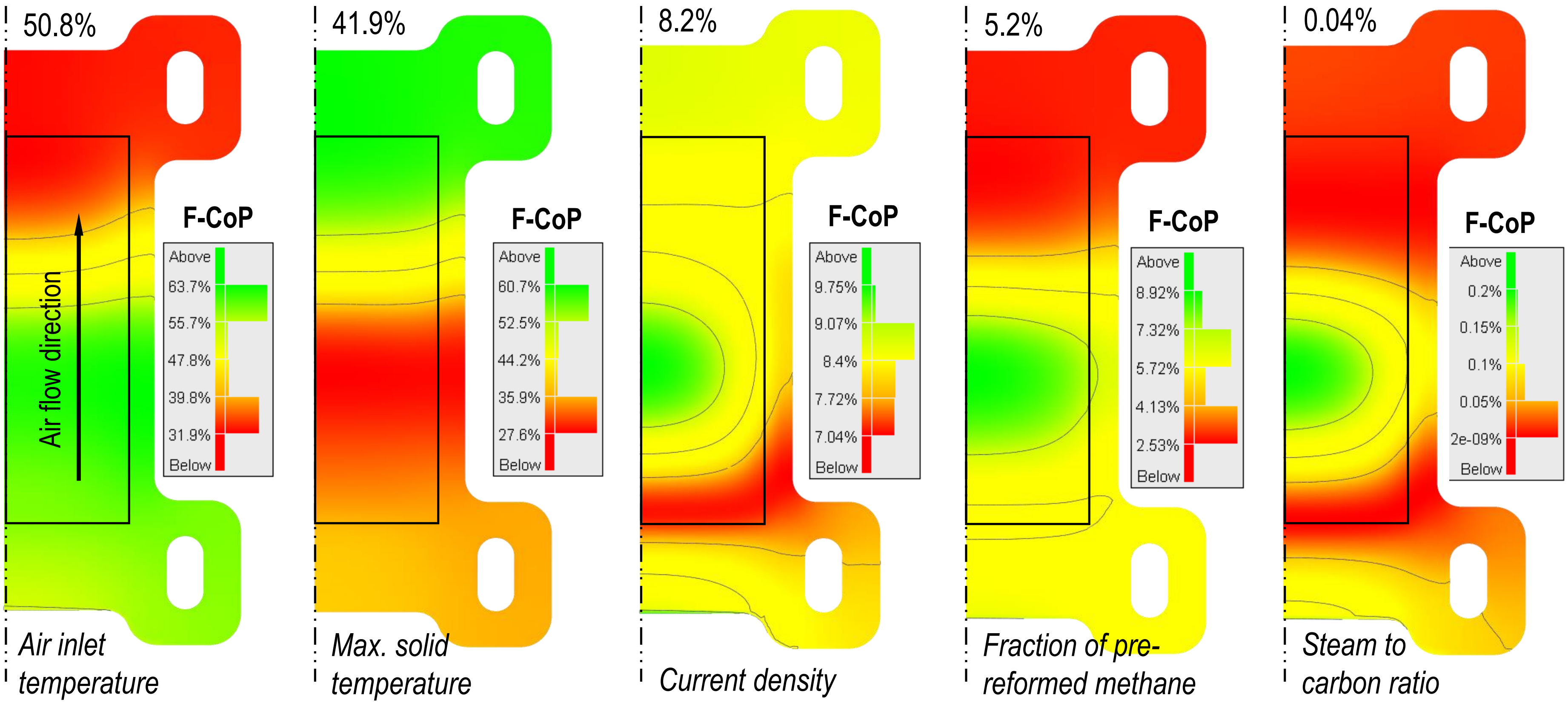
3-D temperature metamodel (3 shapes):



- **Accurate over large operation window.**
- **Achieved local accuracy likely higher than the effects of model simplifications.**

Thermo-electrochemical metamodelling

Temperature profile metamodel: F-CoP for individual factors



Overall expected trends, average effect of pre-reforming less than anticipated.

EPFL SOLID POWER dynardo PROSOFC

Thermo-electrochemical metamodelling

Optimization for spatial temperature control (static analysis)

M. Fardadi, F. Mueller and F. Jabbari, *J. Power Sources* 195 (2010) 4222.

➢ **Minimization of local temperature variations during load following:**

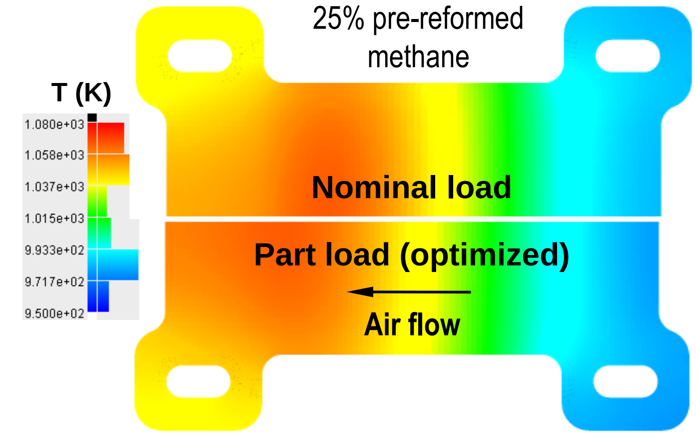
Nominal load (0.35 W cm^{-2})	⟷	**Part load** (0.23 W cm^{-2})

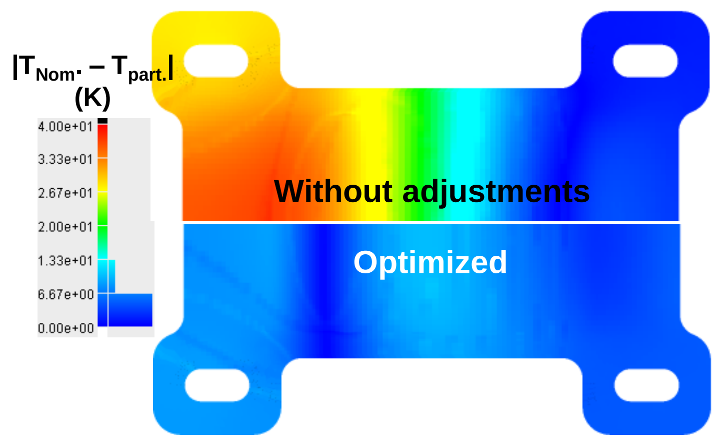
- **Objective function** without region-dependent weighting.
- **Operation conditions manipulated by optimization:**
  - Air inlet temperature.
  - Air ratio (flow).
  - Fuel utilization.

Fraction of pre-reformed methane	$\lvert T_{Nom.} - T_{Part} \rvert_{max.}$ (K)	
	w/o adjustment	Optimized
0.99	**25** (16)	**13** (3)
0.25	**38** (28)	**10** (6)

Parentheses: metamodel calculations

- **Significant reduction of local temperature variations.**
- **Potential further improvements: manipulations of PR.**
- **Constraints from balance of plant components (BoP).**





EPFL SOLID POWER dynardo PROSOFC

Conclusion

Relevance of OptiSLang/SoS for the SOC technology illustrated by 2 examples:

- **Measurement of mechanical properties:**
  - Elastic, **primary** and secondary creep.
  - Numerical and experimental verifications.
  - Application to experimental data for the SOLIDpower Ni(O)-YSZ, wide range of testing conditions.
- **Optimization:**
  - Scalar/**3-D** distributed metamodeling of the stack thermo-electrochemical behavior.
  - Numerical verification and optimization test.
- **Next steps:**
  - Parameter estimation tests with different constitutive laws.
  - Full stack metamodeling, direct comparison with experiments.
  - Implementation of variability in component/assembly quality and defects.

EPFL SOLID POWER dynardo PROSOFC

Acknowledgements

- **PROSOFC consortium**.
- The research leading to these results has received funding from the **European Union's Seventh Framework Programme** (FP7/2007-2013) for the Fuel Cells and Hydrogen Joint Technology Initiative under grant agreement nr. 325278 (Project name: **PROSOFC**), funding for the Swiss partners was provided by the **Swiss Federal Office of Energy** under contract nr. SI/500084-04.