National Research and Development Institute for Cryogenics and Isotopic Technologies - ICSI Rm. Valcea, Romania

PEM Fuel Cells
- On the Way from Modelling to Reality

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Conference on Energy, Environment, Economy and Thermodynamics, 14-15 october, Bucharest

OVERVIEW

- ICSI RM. VALCEA – RESEARCH DIRECTIONS AND ACHIEVEMENTS
- PEM FUEL CELLS – A KEY ELEMENT IN OUR RESEARCH ACTIVITIES
- MATHEMATICAL MODEL DEVELOPMENT
- RESULTS & DISCUSSIONS
- FUTURE WORK & CONCLUSIONS

ICSI

The National Research & Development Institute for Cryogenics and Isotopic Technologies – ICSI Rm. Valcea was founded in 1970 as the "G“ Plant, being an experimental industrial pilot plant for heavy water production.

ICSI - an organization on three pillars

ICSI NUCLEAR

To develop research in the field of nuclear fission, nuclear fusion, isotopic exchange and cryogenic distillation

ICSI ENERGY

To develop and implement hydrogen-based technologies and renewable energy sources

ICSI ANALYTICS

To implement appropriate analytical methods and markers for environmental protection and food security

ICSI Energy – on the road towards hydrogen economy

- Working with hydrogen since the 90s

National Center for Hydrogen and Fuel Cell – 2009

Low Temperature Laboratory for energy support – 2012

ROManian **E**nergy **ST**orage Laboratory - 2015

Objectives:

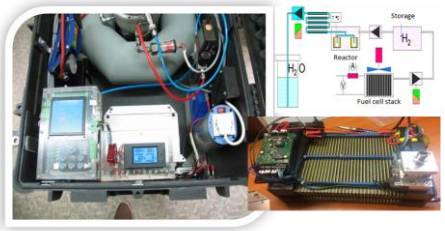
- Promote excellence in fundamental and applied research
- Provide support for development of applied technologies and models
- Advice and support for authorities regarding RES and hydrogen
- Support in training activities for students and young researchers

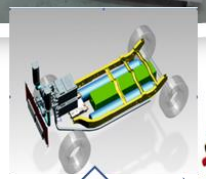
- **2000** – R&D projects in the field of H2 and fuel cells for energy applications
- **2009** – establisment of National Center for Hydrogen and Fuel Cells
- **2009** – ICSI Energy became a full member of the **Joint Technology Initiative – Joint Undertaking on Hydrogen and Fuel Cell**, a public-private partnership supporting RD&D in the field of fuel cells and hydrogen
- **2012** – foundation of the **Romanian Association for Hydrogen Energy**
- **2014 –** ICSI Energy became a National Interest Facility
- **2021** - ICSI Energy became a member of the European Clean Hydrogen Alliance

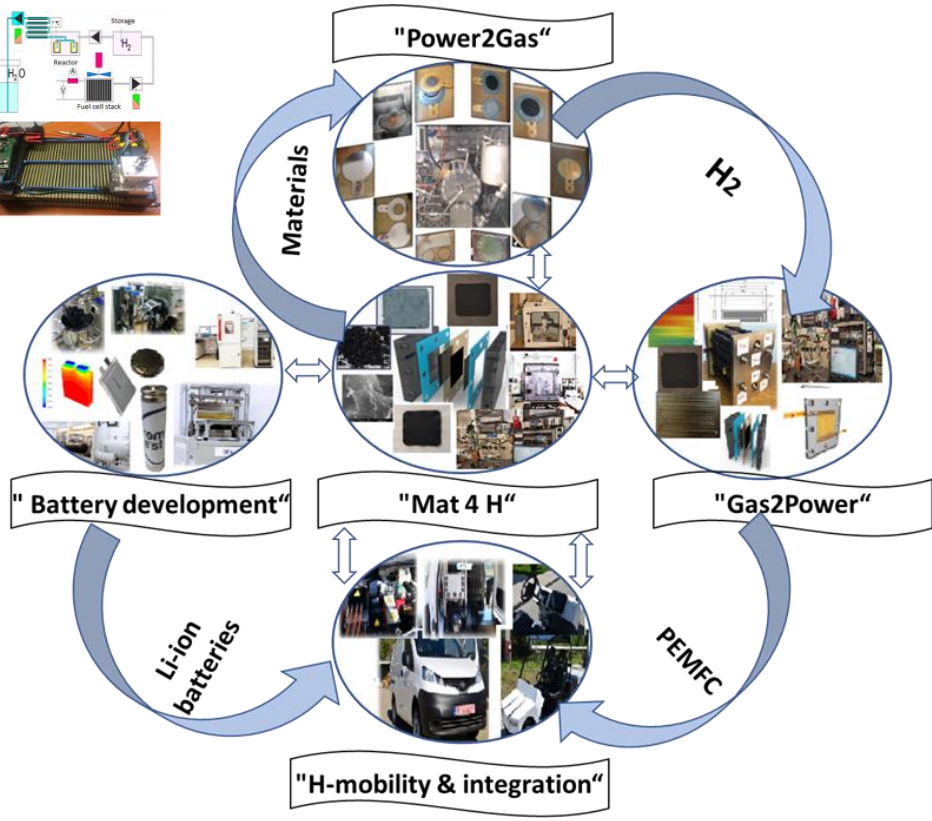
ICSI Energy – main research directions and achievements

- Development of new materials for electrochemical devices investigated-"Mat 4 H“
- Conversion of hydrogen into energy using fuel cells - "Gas2Power".
- Conversion of energy into hydrogen using electrolysis - "Power2Gas“.
- Development of hybrid energy storage technologies - "Lithium-Ion-Battery".
- Development of "clean" mobility engines - "H-mobility".

Stationary CHP (170cm^2-2kW).

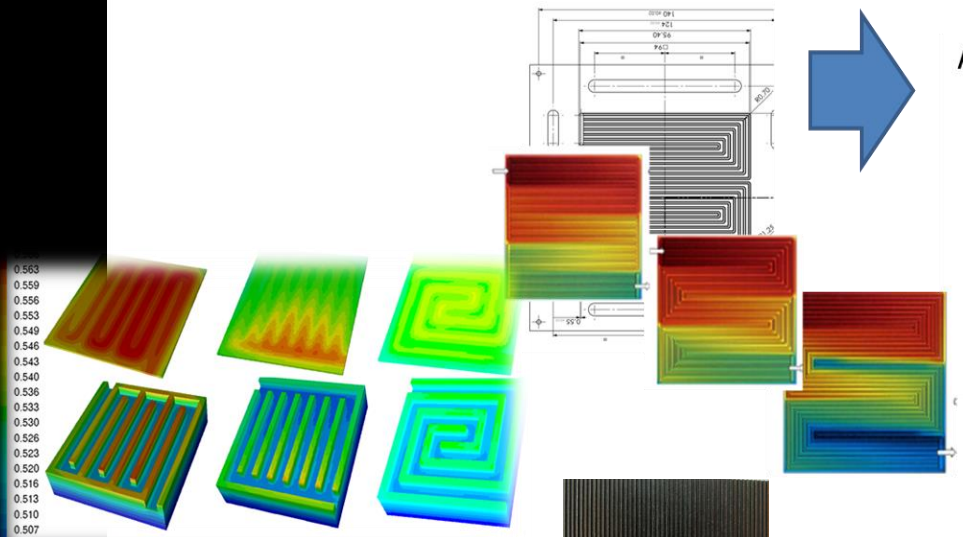






"Gas2Power" program - "From modelling to reality"

- Design of PEM fuel cells and assemblies of fuel cells
- Development of PEMFC components: bipolar and collector plates, MEA assemblies, microporous and gas diffusion layers, catalysts
- Integration of components into optimized configurations from volumetric and generated power point of view
- Electrical/electrochemical testing of PEM fuel cells and assemblies
- Integration in educational systems (25cm^2-25W), portable (44cm^2-250W), stationary / back-up off grid (92cm^2-1kW).

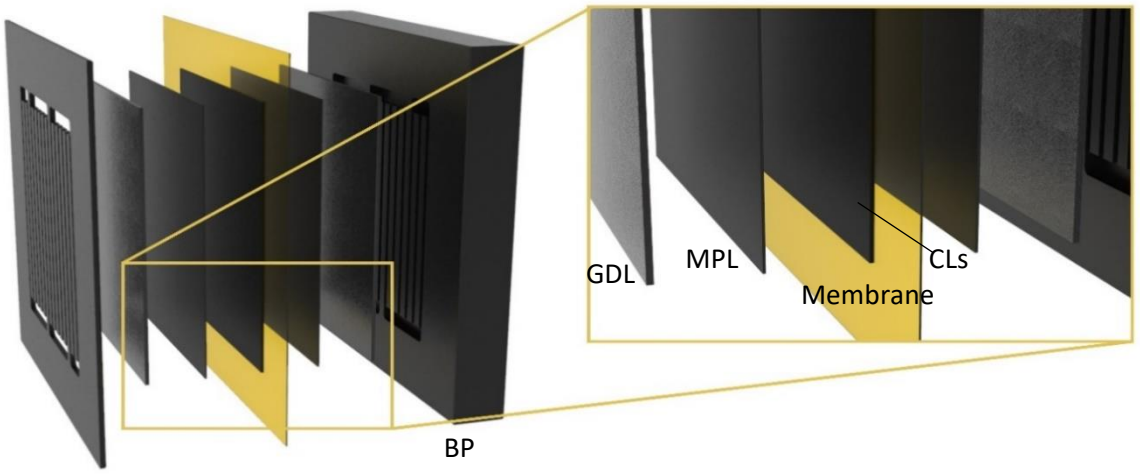


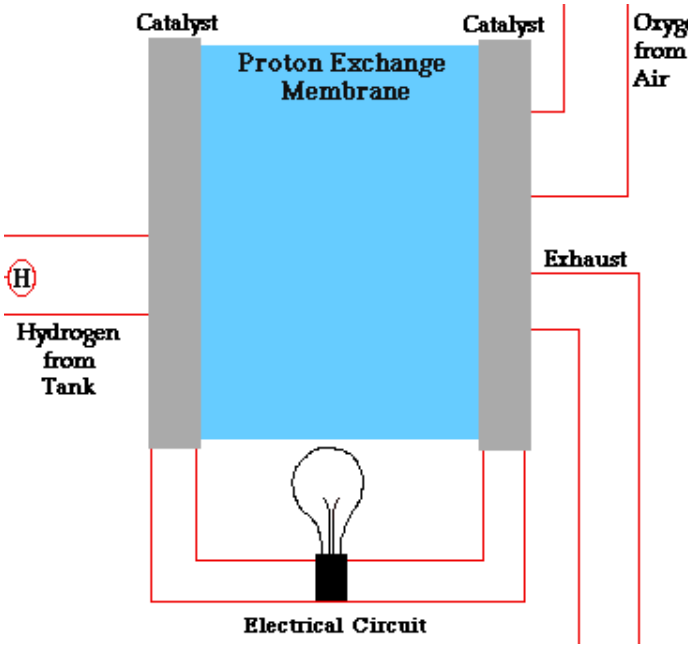
ANSYS + FUEL CELL MODULE
COMSOL

PEM FUEL CELLS

– ONE KEY ELEMENT IN OUR RESEARCH ACTIVITIES

PEM FUEL CELL DEFINITION





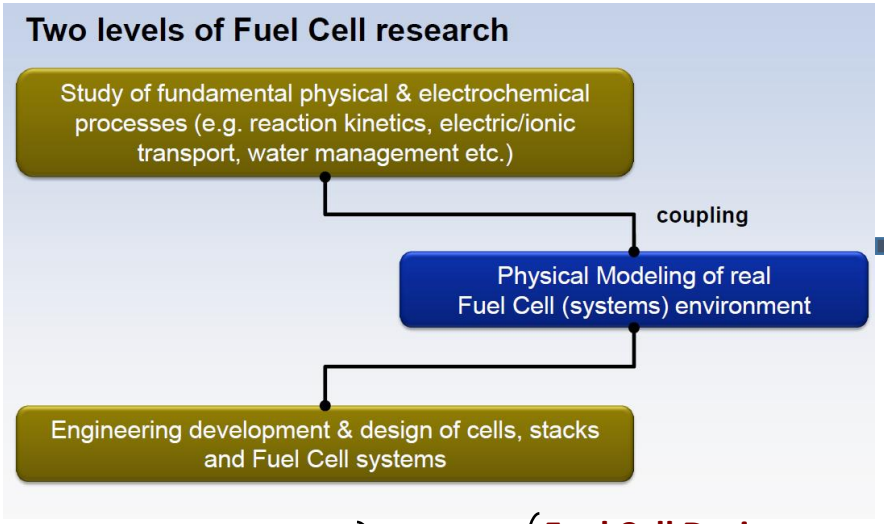
A Fuel Cell is an electrochemical device that combines hydrogen and oxygen to produce electricity, with water and heat as by-products.

ANODE: $H_2 \rightarrow 2H^+ + 2e^-$

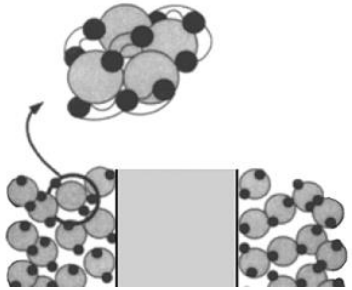
CATHODE: $\frac{1}{2}O_2 + 2H^+ + 2e^- \rightarrow H_2 0$

OVERALL REACTION: $2H_2 + O_2 \Rightarrow 2H_2O + energy + heat$

Motivation and Issues for PEM Modeling



- Large variation of important length scales
  - Electrochemistry at nm
  - Porous Flow at um
  - Flow Fields at mm
  - Balance of Plant at m



- Combination of fundamental processes
  - Electrochemistry and Water transport
  - Flow and Heat generation and transport
  - Charge transport, diffusion and reaction
- Complexity of geometrical structure
  - Layered structures
  - Repeated sub systems

Requirements

- **Power Density**
- **Efficiency**
- **Start-up**
- **Transient response**
- **Weight**
- **Size/shape**

Operation Conditions

- **Temperature**
- **Fuel and Oxidant**
  - Composition/Purity
  - Pressure
  - Temperature
  - Humidity
  - Flow Rate

Fuel Cell Design

- **Bipolar plate**
  - Channel size
  - Shoulder size
  - Channel layout
- **Gas Distribution Electrode**
  - Thickness 100-300 µm
  - Porosity
  - Conductivity
- **Membrane**
  - Thickness 50-250 µm
  - Type
- **Catalyst Layer**
  - Thickness 10-30 µm
  - Composition

PEM FUEL CELLS CFD MODELING using ANSYS Fluent

- PEM Fuel cell modeling requires a proper description of:
  - Fluid flow and heat & mass transfer in both gas channels and porous electrodes
  - Electrochemical reactions
  - Multiphase flow with phase change
  - Transport of current and potential field in porous media and solid conducting regions.

Several assumptions are made in developing the model :

- ❖ *the fuel cell operates under steady state conditions*
- ❖ *the fuel and the air flows can be treated as an ideal gas mixture*
- ❖ *the flow in the channels is considered incompressible and laminar*
- ❖ *the cell operates under non-isothermal conditions.*

GOVERNING EQUATIONS

Mass conservation

$$\nabla \cdot \left(\varepsilon \rho \vec{\mathrm{u}}\right) = \mathrm{S_m}$$

$$S_m = \begin{cases} -M_{H_2O} \cdot \alpha - M_{H_2} \cdot \dfrac{R_{anode}}{2F} & in \quad anode \\ M_{H_2O} \cdot \dfrac{R_{cathode}}{2F} + M_{H_2O} \cdot \alpha - M_{O_2} \cdot \dfrac{R_{cathode}}{4F} & in \quad cathode \end{cases}$$

Momentum conservation

$$\nabla \cdot \left(\varepsilon \rho \vec{u}\vec{u}\right) = -\varepsilon \nabla p + \nabla \cdot \left(\varepsilon \mu \nabla \vec{u}\right) + S_u \qquad S_u = -\frac{\mu}{k} u$$

Species conservation

$$\nabla \cdot \left(\varepsilon \vec{u} Y_i\right) = \nabla \cdot \left(D_i \nabla Y_i\right) + S_i$$

$$S_{H_2} = -\frac{M_{H_2}}{2F} \cdot R_{anode}$$

$$S_{O_2} = -\frac{M_{O_2}}{4F} \cdot R_{cathode}$$

$$S_{H_2O} = \begin{cases} -M_{H_2O} \cdot \alpha & in\ the\ anode\ catalyst\ layer \\ M_{H_2O} \cdot \alpha + \dfrac{M_{H_2O}}{2F} \cdot R_{catod} & in\ the\ cathode\ catalyst\ layer \end{cases}$$

Charge conservation

$$\nabla \cdot \left(\sigma_e \nabla \phi_e\right) + S_\phi = 0$$

$$S_{\phi_s} = \begin{cases} -R_a & in\ anode\ catalyst\ layer \\ R_c & in\ cathode\ catalyst\ layer \end{cases}$$

$$S_{\phi_e} = \begin{cases} R_a & in\ anode\ catalyst\ layer \\ -R_c & in\ cathode\ catalyst\ layer \end{cases}$$

Energy conservation

$$\left(\rho \cdot c_p\right)\left(\vec{u} \cdot \nabla T\right) = \nabla \cdot \left(k_{eff} \nabla T\right) + S_h \qquad S_h = h_{react} + h_{phase} + I^2 R_{ohm}$$

With and without mass transport resistance due to catalyst microstructure

$$R_{cat} = 4F \frac{c_{O_2}}{\left({c_{O_2}}/{j_{O_2}^{ideal}} + R_{ion} + R_{liq} \right)}$$

$$j_{O_2}^{ideal} = {R_{cat}^{0}}/{4F}$$

$$R_{cat}^{0} = \left(\zeta_{cat} \cdot j_{cat}^{ref}\right) \cdot \left(\frac{c_{O_2}}{c_{O_2}^{ref}}\right)^{\gamma_{cat}} \cdot \left[-e^{\frac{\alpha_{an} F \eta_{cat}}{RT}} + e^{\frac{-\alpha_{cat} F \eta_{cat}}{RT}}\right]$$

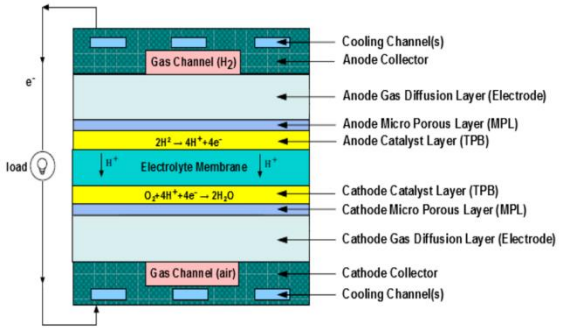
$$R_{liq} = \left(\frac{\zeta_{cat} \cdot r_p^2}{K_w \cdot D_w}\right) \cdot \frac{\left(\sqrt[3]{1 + \frac{s\varepsilon}{1-\varepsilon}} - 1\right)}{3 \cdot (1-\varepsilon)}$$

New model – catalyst microstructure influence

R_{ion} is the resistance due to ionomer film= constant from experiments

$$R_{cat}^{0} = \left(\zeta_{cat} \cdot j_{cat}^{ref}\right) \cdot \left(\frac{c_{O_2}}{c_{O_2}^{ref}}\right)^{\gamma_{cat}} \cdot \left[-e^{\frac{\alpha_{an} F \eta_{cat}}{RT}} + e^{\frac{-\alpha_{cat} F \eta_{cat}}{RT}}\right]$$

Old model – homogeneous structure



Steps of Numerical Investigation

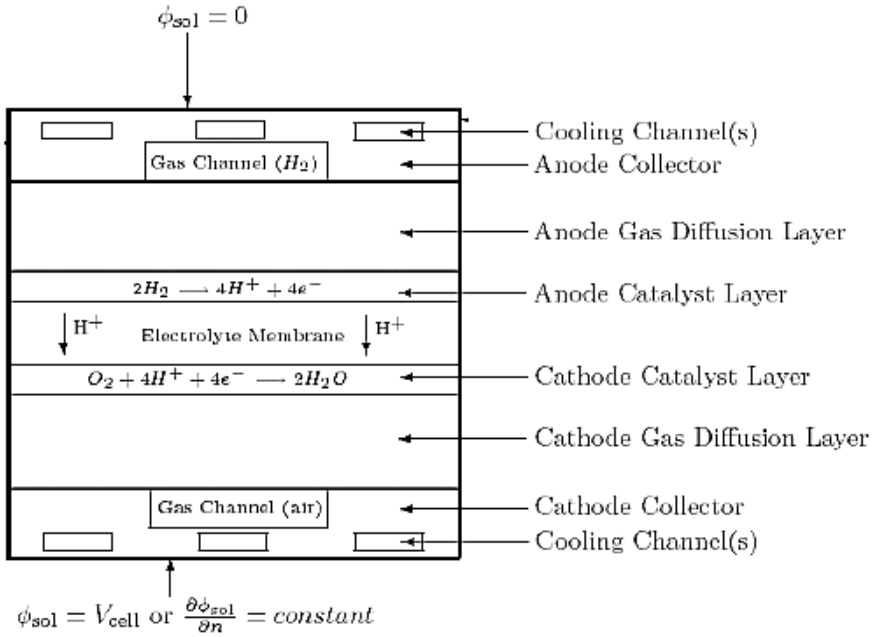
- Create/import the model geometry
- Develop mesh (computational domain)
- Define boundary conditions
- Set up the most appropriate CFD model (Steady state or Transient, isothermal or non-isothermal investigation, incompressible or compressible fluid flow
- Initialize solution and run iterations
- Obtain and plot results.

Specific to fuel cell simulations:

-Potentiostatic or galvanostatic boundary conditions are applied on the external contact boundaries for the solid phase potential, ϕ_{sol}.

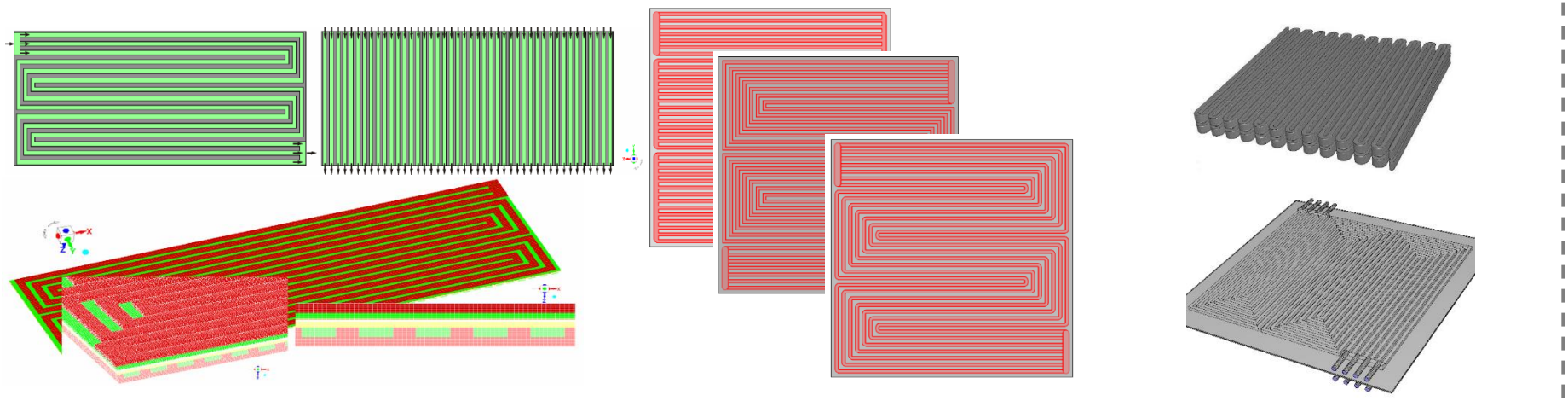
BOUNDARY CONDITIONS

	Parameter	Value	Unit
Anode inlet	Mass flow rate	8e-6	kg/s
	Mass fraction for H_2	0.554	-
	Mass fraction for H_2O	0.446	-
Cathode inlet	Mass flow rate	1.1e-4	kg/s
	Mass fraction for O_2	0.215	-
	Mass fraction for H_2O	0.053	-
Operating conditions	Anode/cathode potential difference	0.4-0.8	V
	Temperature	338	K
	Pressure	152.000	Pa



Bipolar plate and channels design investigation – what we did

Finding a flow field pattern that distribute the gas more evenly is one method in minimizing some problems that appear in PEM fuel cells and can lead to optimization of the PEM fuel cell performance.



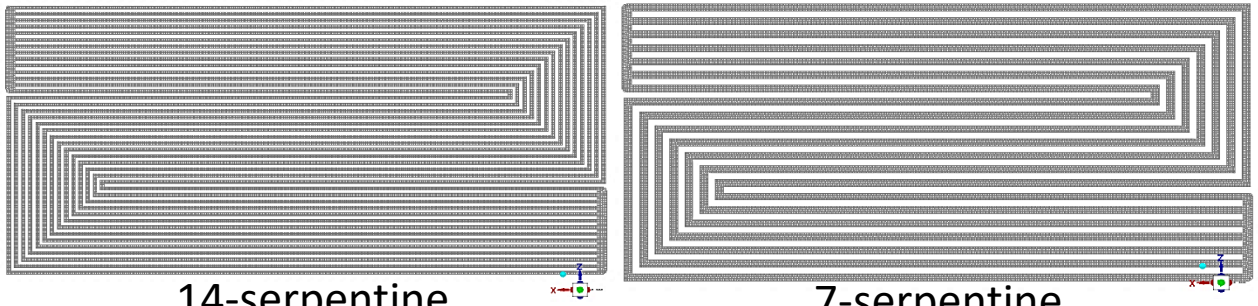
Have been investigated:

- Different bipolar plate sizes: from 5 up to 100 cm^2
- Different configurations for anode and cathode
- Various flow field patterns: serpentine, parallel, mixed

Bipolar plate and channels design investigation – what we are doing and will do

We moved to large scale PEMFCs which require optimized designs

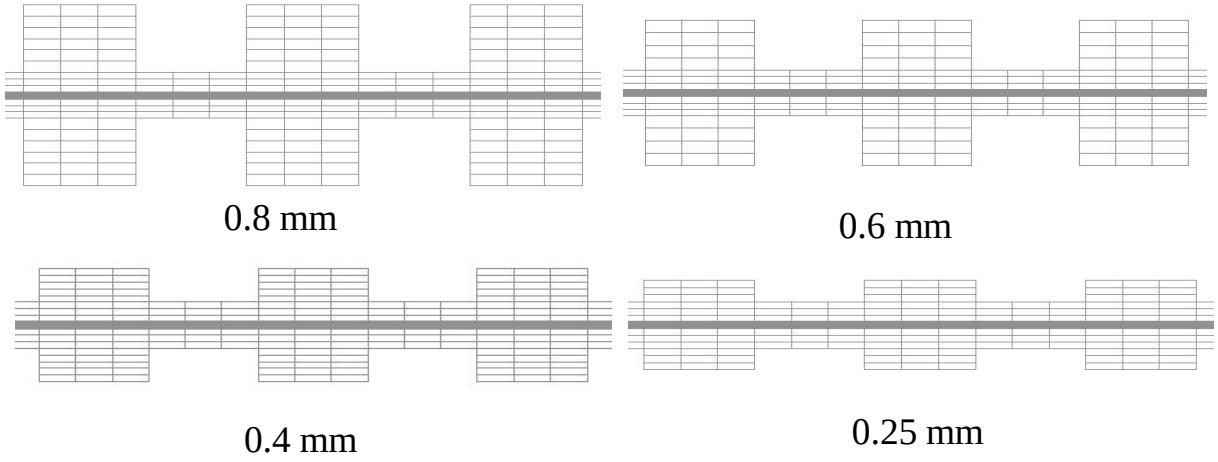
1. Determine the best flow field pattern



14-serpentine 7-serpentine

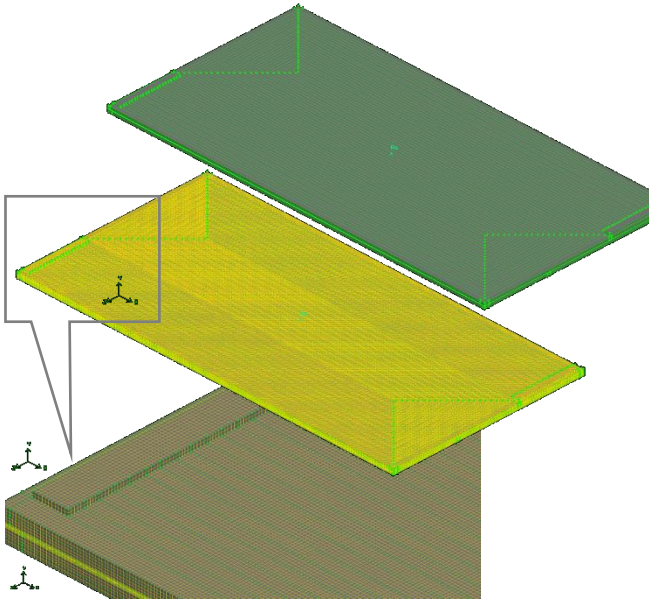
170 cm^2 active area

2. Determine the optimum flow field depth



Channel width 1.3 mm
Land width 1.4 mm

Computational geometry with mesh



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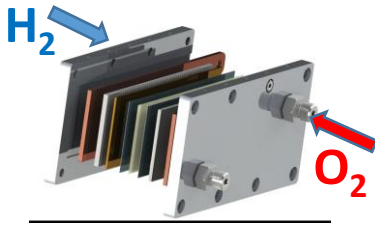
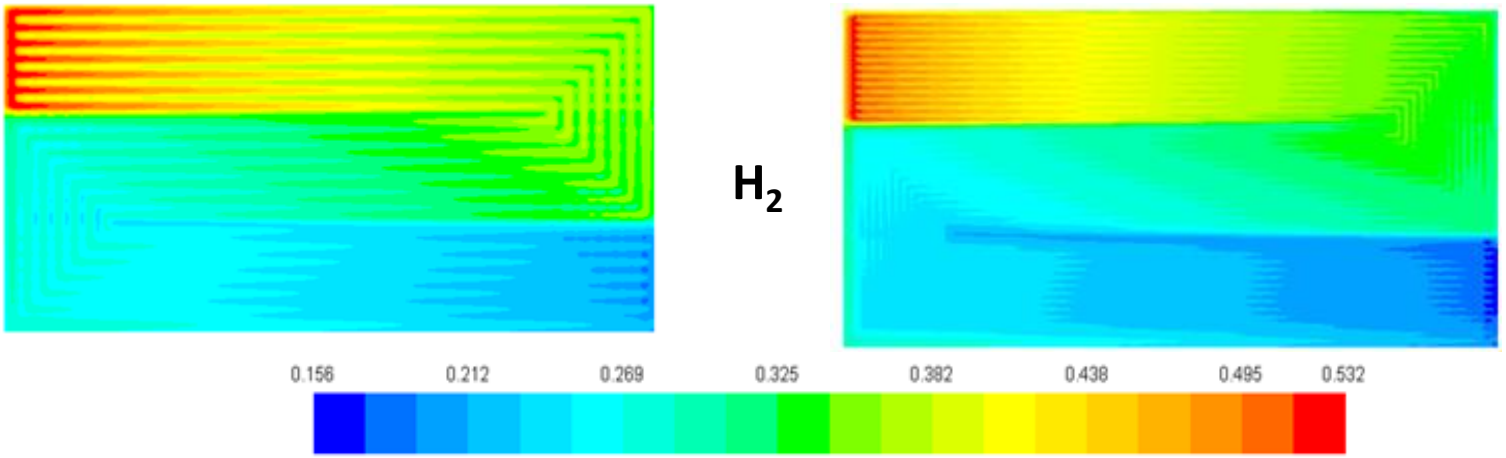
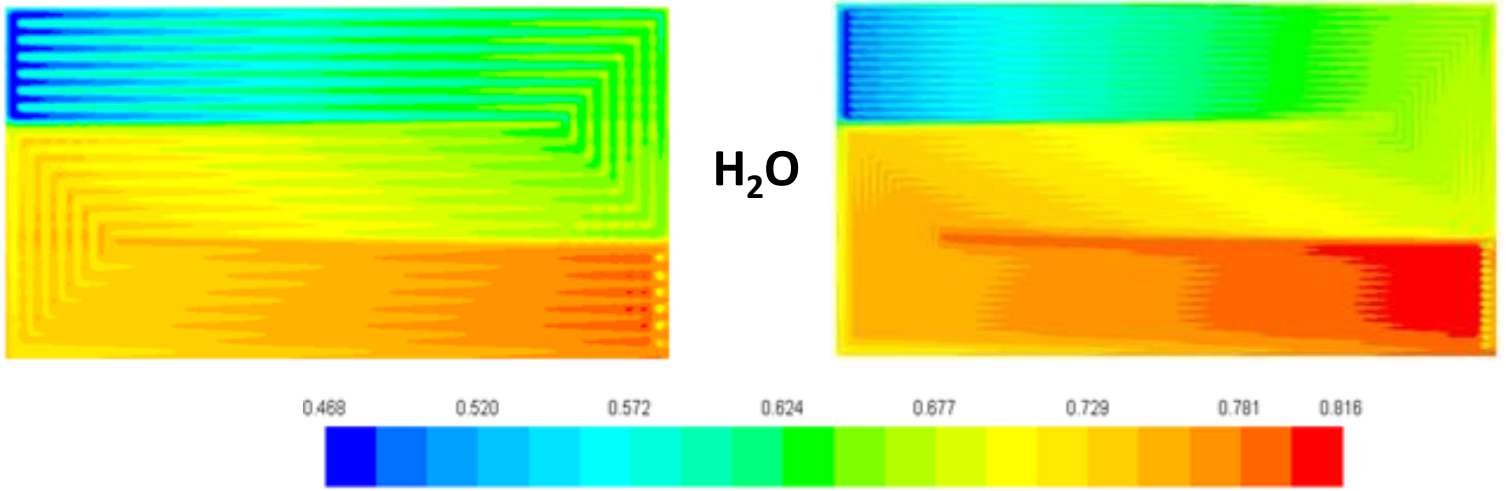


- Hydrogen and water mass fraction contours in anode catalyst layer



7-serpentine channel

14-serpentine channel



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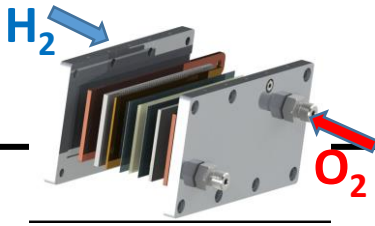
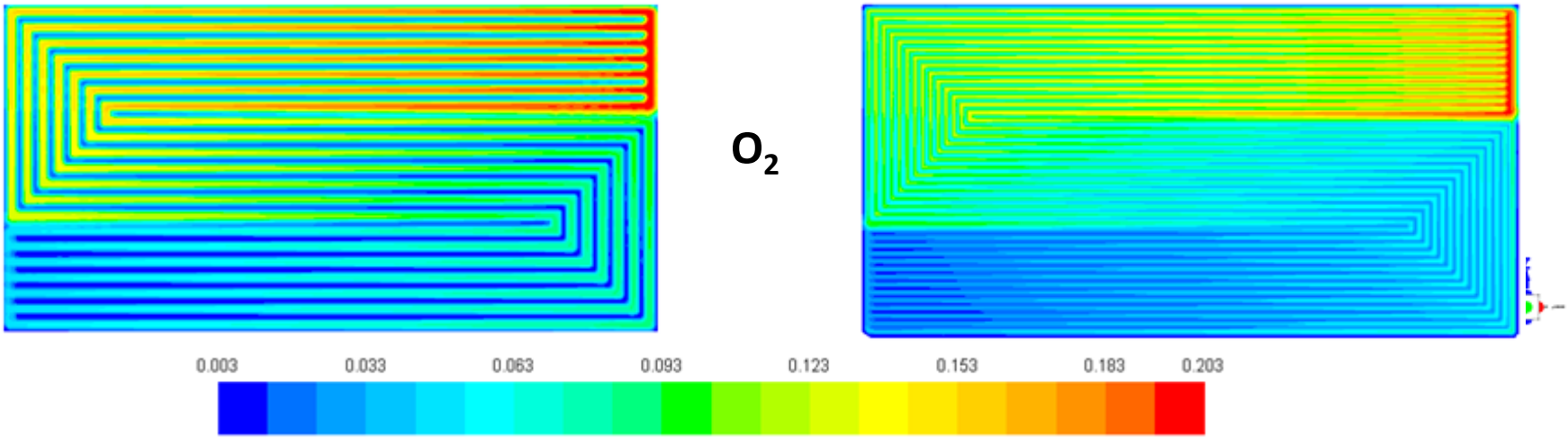


- Oxygen and water mass fraction contours in cathode catalyst layer



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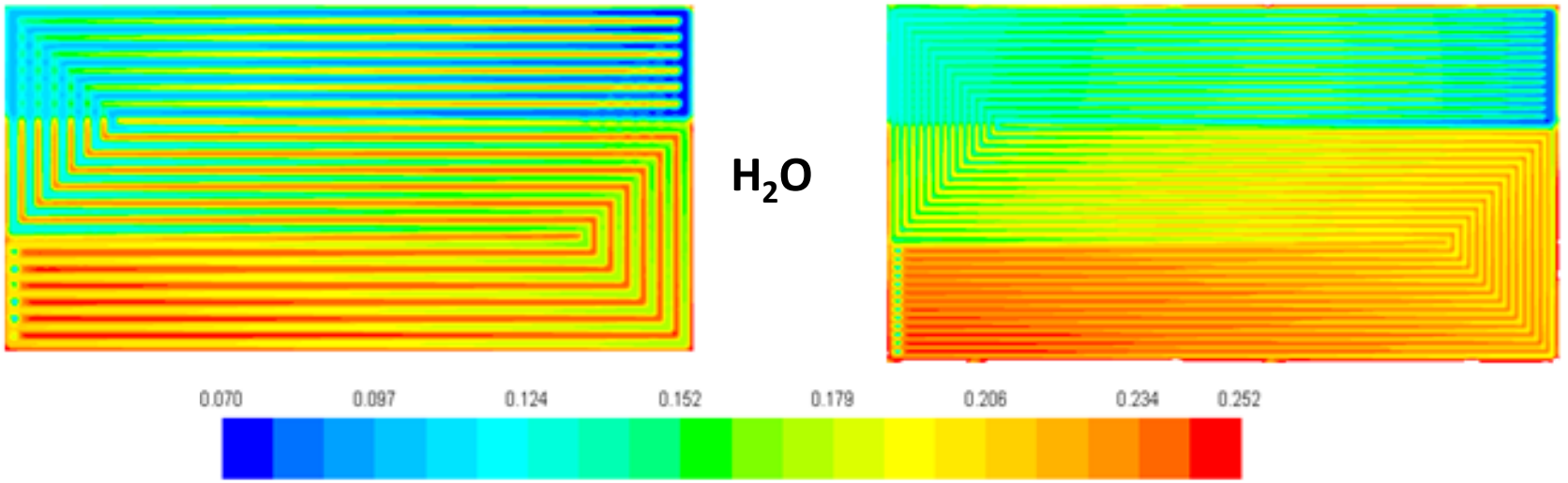
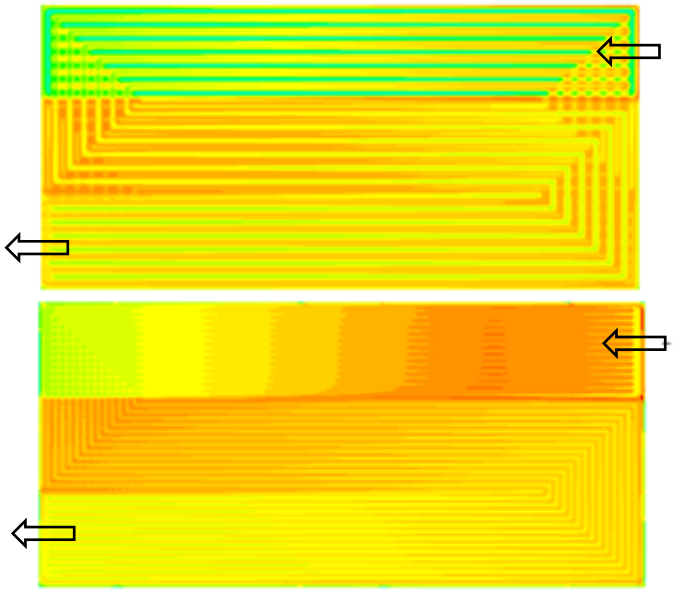


Water content in membrane (λ) at 0.6V



7-serpentine channel

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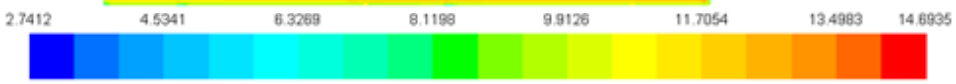
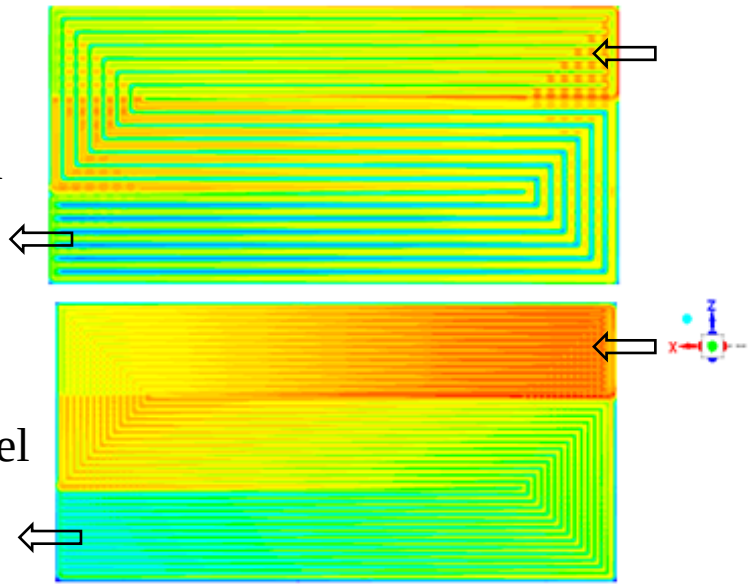


Current density (A/m^2) at 0.6V

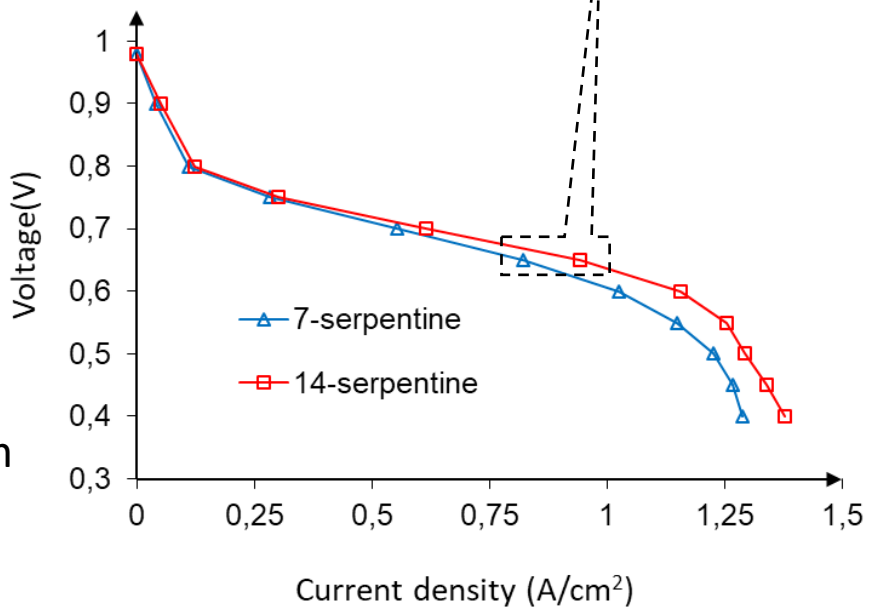


$$\lambda = \begin{cases} 0.043 + 17.18 \cdot a - 39.85a^2 + 36.0a^3 & \text{for} \quad 0 < a \le 1 \\ 14 + 1.4 \cdot (a - 1) & \text{for} \quad 1 \le a \le 3 \end{cases}$$

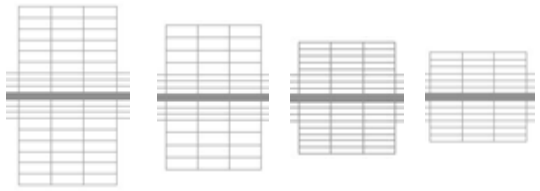
$$a = \frac{X_{H_2O} p}{p^{sat}}$$

⇨ 5% increase in performance at low voltage for 14-serpentine channel fuel cell

⇨ 14-serpentine channel fuel cell is the optimum design and it is used for further investigations

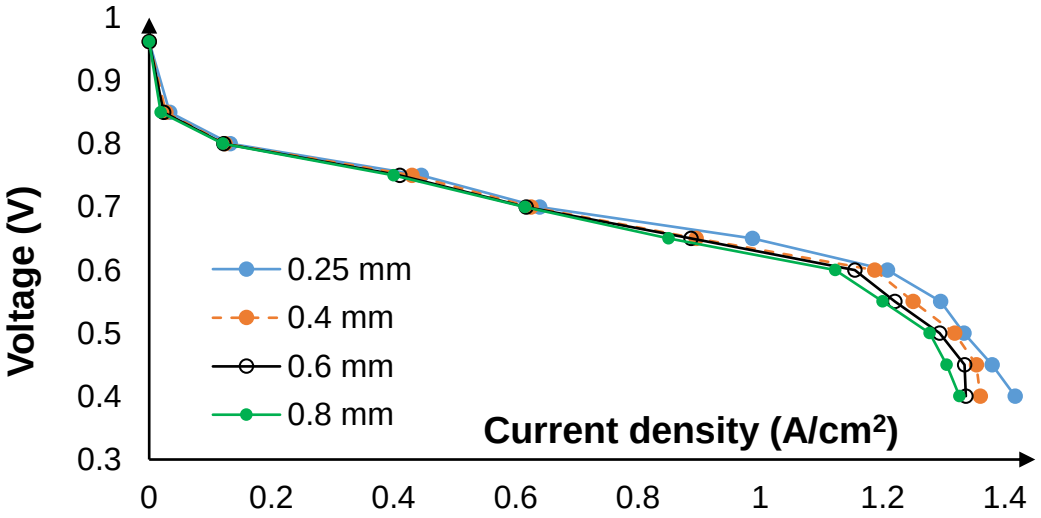


2. Determine the optimum flow field depth



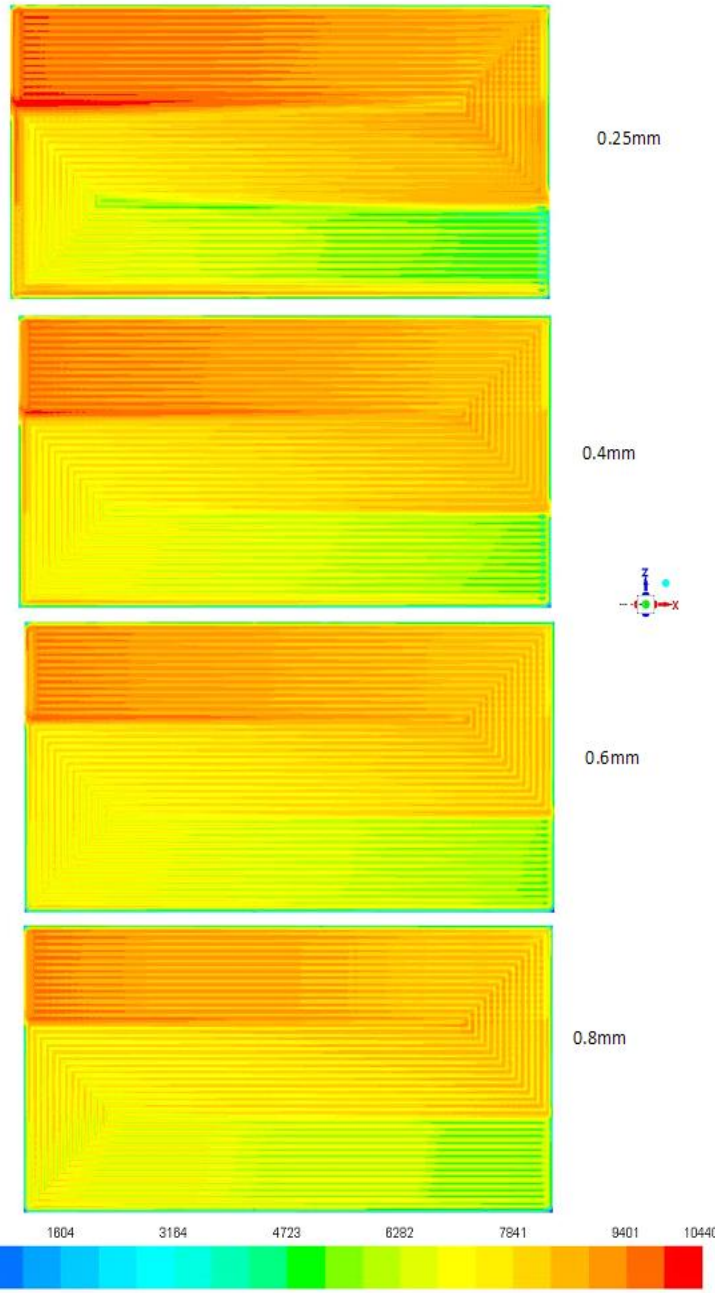
0,8 0,6 0,4 0,25

mm

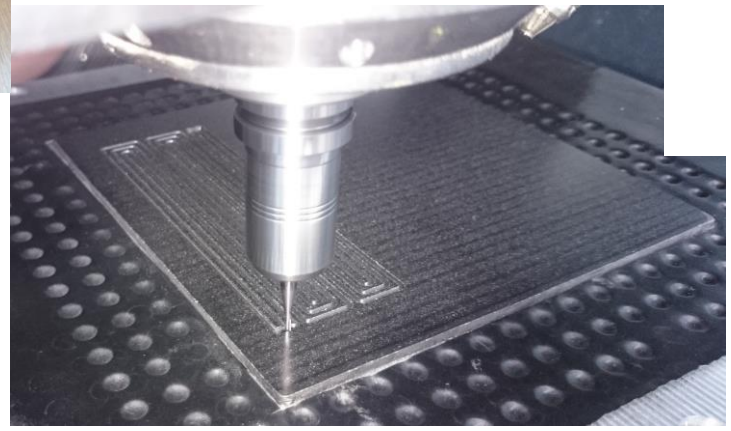


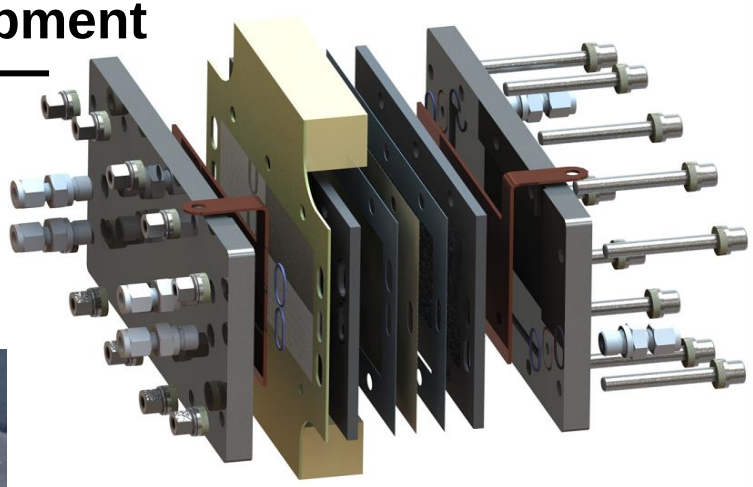
⇨ fuel cell performs better as the channel depth decreases (7% increase in performance at 0.4V)

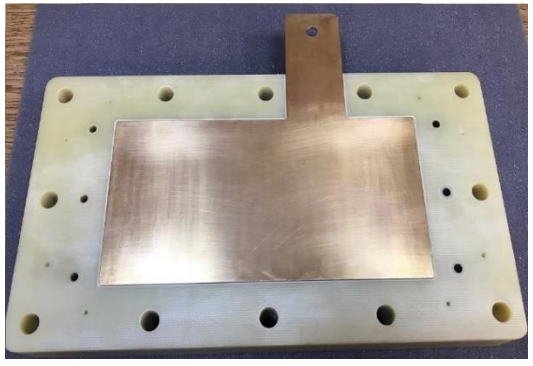
The non-uniform current density distribution may cause local hotspots, thus affecting the longevity and durability of the fuel cell.

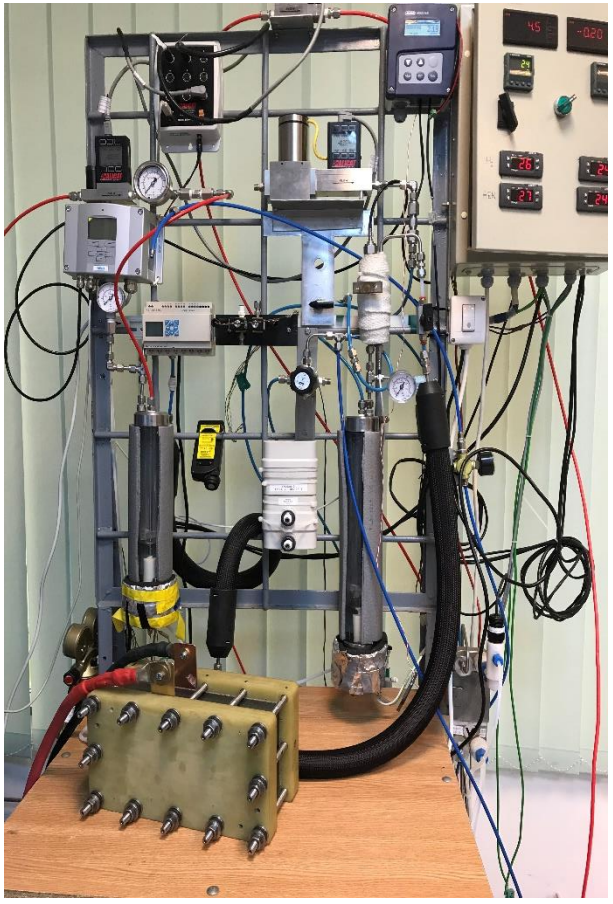


Bipolar plates experimental development

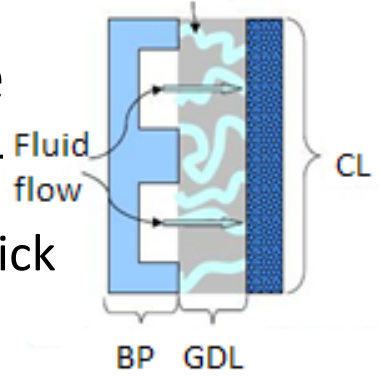








2. Influence of GDL on the PEMFC performance

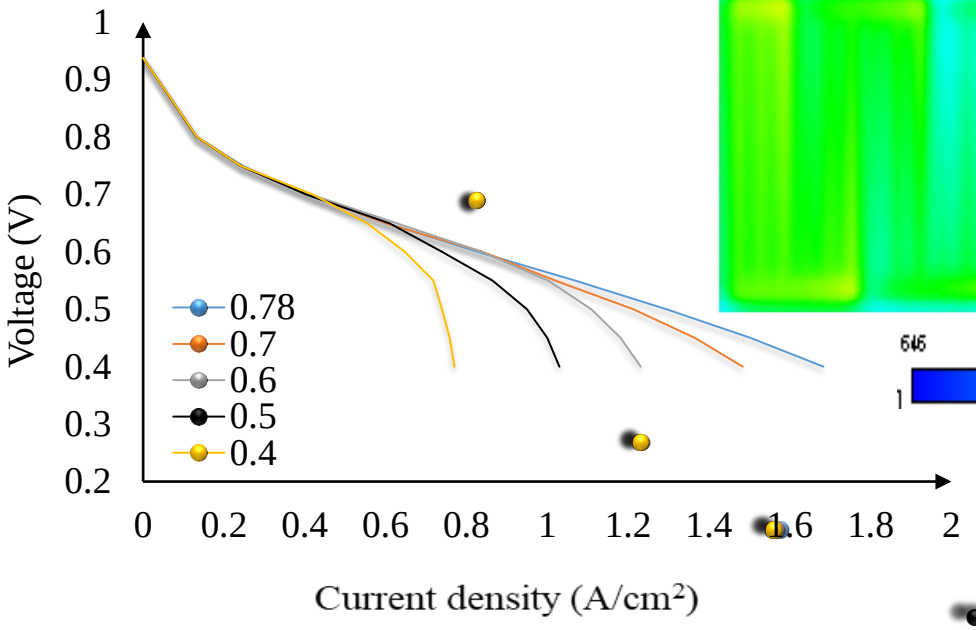


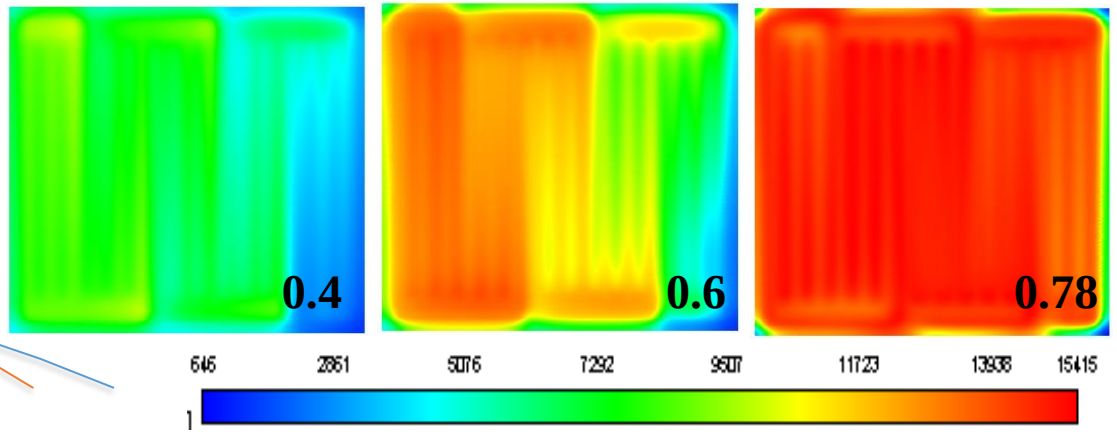
- GDL is made typically on carbon paper or carbon cloth, ~0.3 mm thick
- GDL:
  - provide passages for reactant transport and heat/water removal
  - provide an electrically conductive pathway for current collection
  - provide mechanical support to the MEA.

- PEMFC performance is mainly influenced by

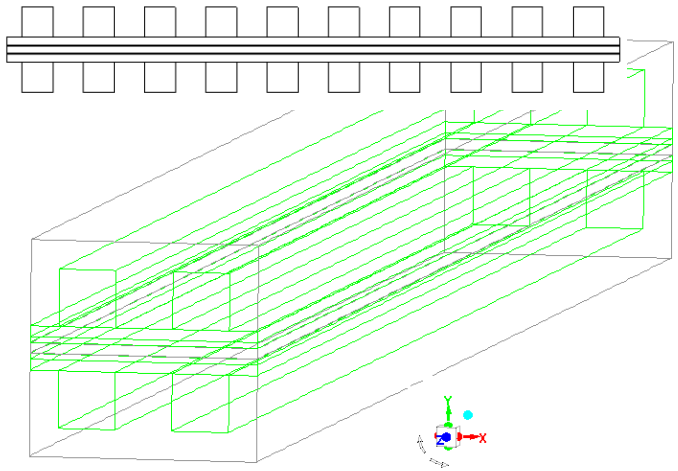
  GDL porosity ↔ GDL permeability ↔ GDL compression

- Higher gas diffusion layer porosity improves the mass transport within the cell.





GDL COMPRESSION



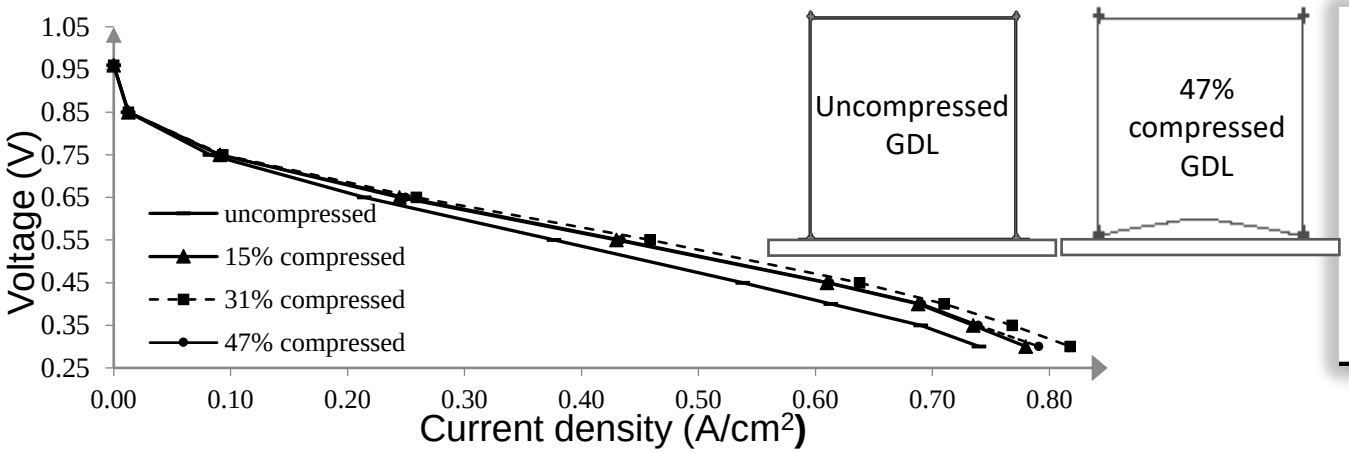
- Gas Diffusion Layer, the most deformable component of the PEM fuel cell, can deform differently under the applied compression.

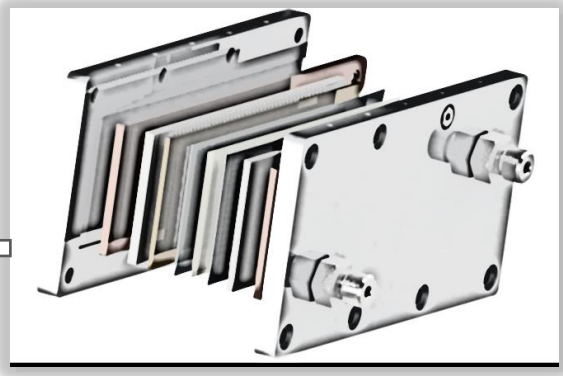
- Under the land of the BP → compression applied modify the GDL thickness, porosity, hydrophobicity and electrical resistance
- Under the channels → the GDL may intrude in the flow field channels, leading to significant disturbance of the reactants flow in the channels.

Compression influences *mechanical, electrical and chemical properties of fuel cells*. ***Optimal fuel cell compression must be determined*** for maximizing performance, increasing durability and life time.

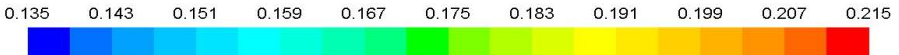
- excessive compression can cause mechanical damage of the fuel cell components and gas blockage
- insufficient compression can leads to an increase of contact resistance between layers and to gas leakage

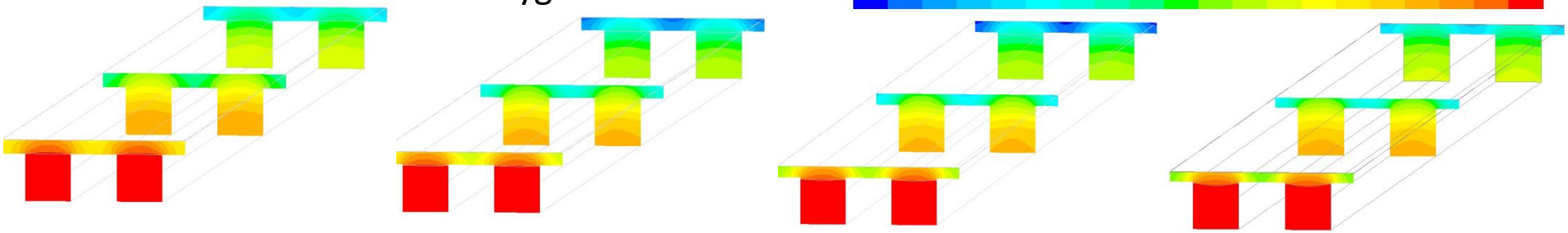
INVESTIGATION OF COMPRESSION ON THE PEMFC PERFORMANCE



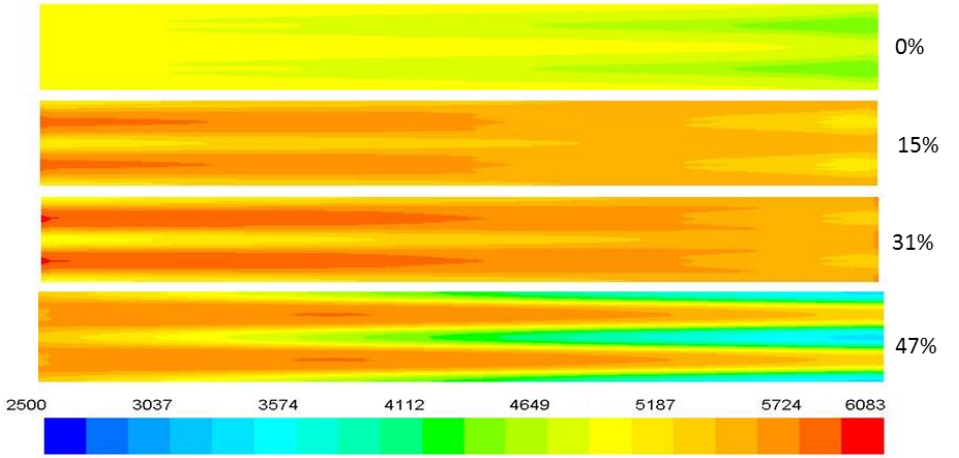


Oxygen mass fraction

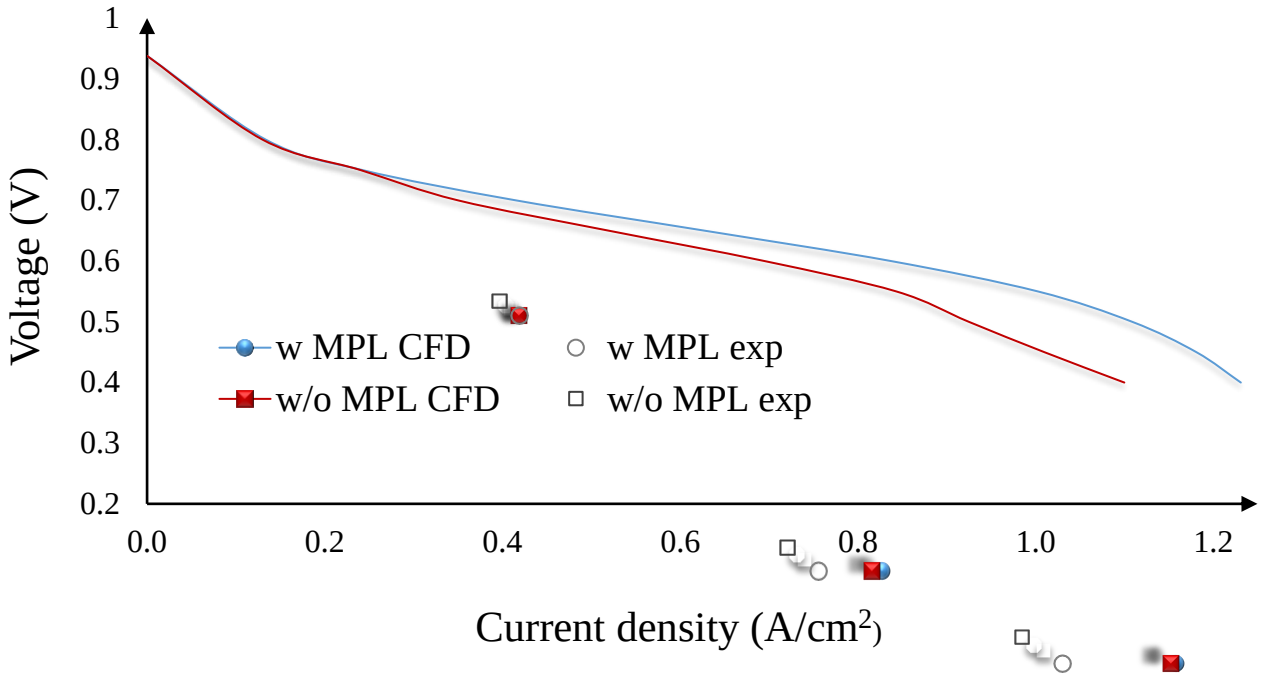




- As the compression increases- the cell performance improves due to the reduced interfacial resistance.
- A bigger compression leads to a lower porosity of the GDL (less void space for the reactants/products to pass through) and consequently a decrease in the fuel cell performance.

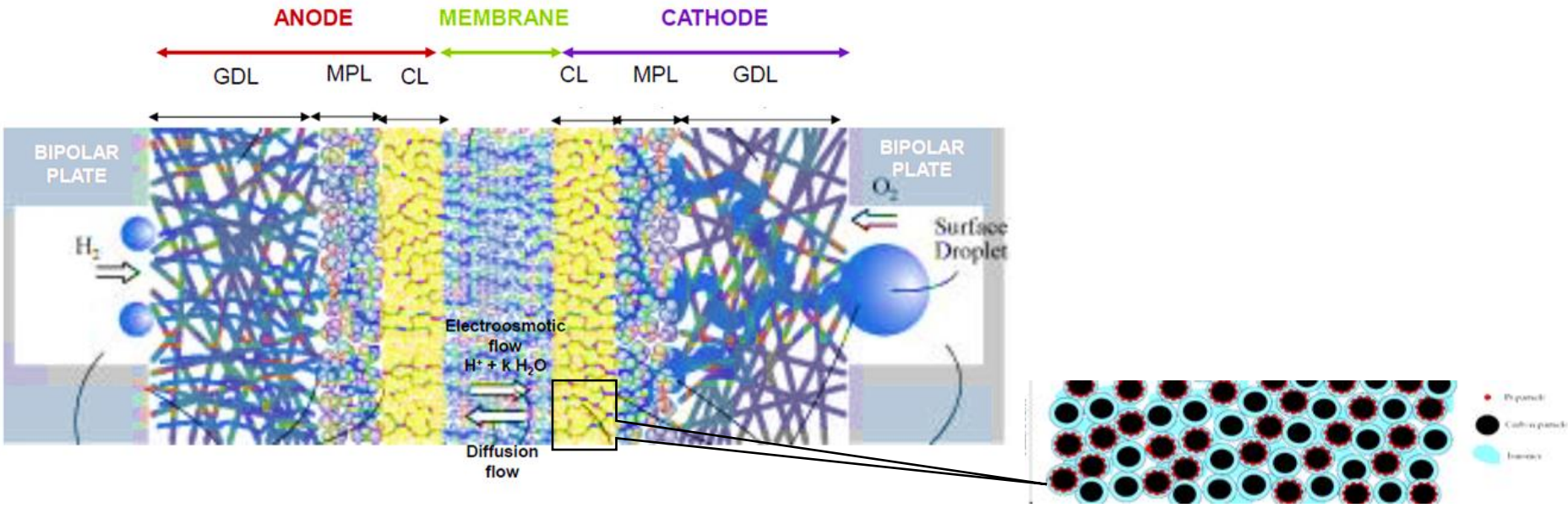


3. MICRO POROUS LAYER INVESTIGATION



- An improved performance of the fuel cell is obtained when the MPL was taken into consideration in the ohmic and concentration polarization area.
- The dense microstructure of MPL helps in reducing the contact resistance between GDL and CL and pushes liquid water generated from cathode side back to the anode side hydrating the membrane.

4. CATALYST LAYER MICROSTRUCTURE INVESTIGATION

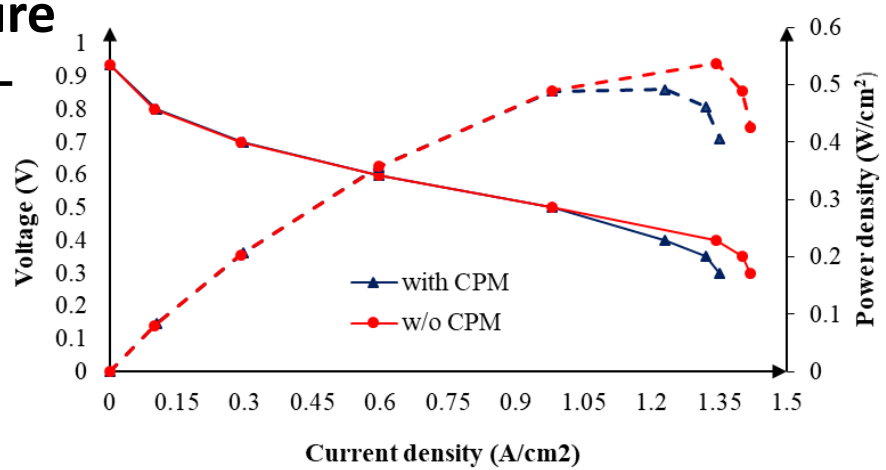


The catalyst layer must provide

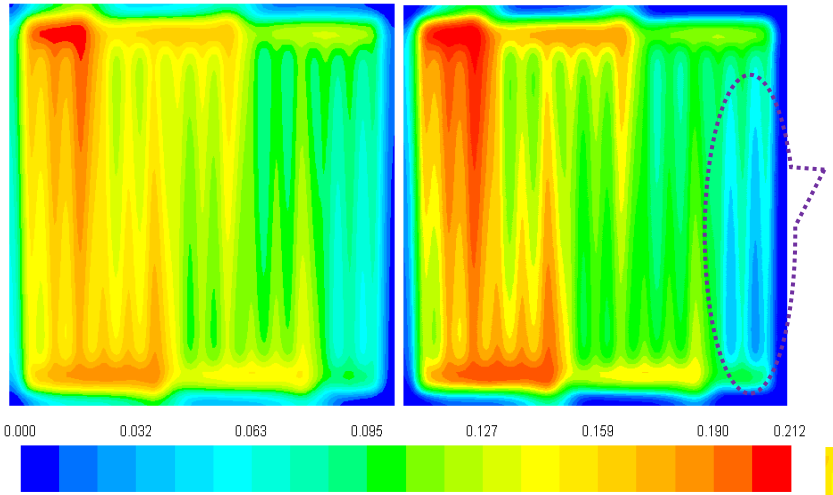
- channels for the transport of reactants and products,
- electrically conductive path for the transport of electrons and
- an ion conductive path for the transport of protons from the electrode to the membrane.

The electrode must have a balance in order to avoid performance losses and maximize the utilization of the Pt base catalyst.

With and without CL microstructure



O_2 mass fraction profile at 0.3V

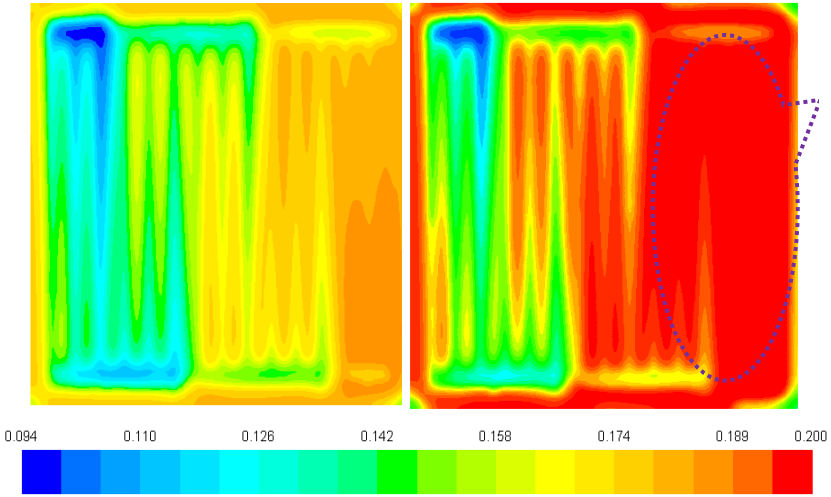


without CL microstructure

with CL microstructure

Depletion of oxygen captured by the CPM model

Water mass fraction profile at 0.3V



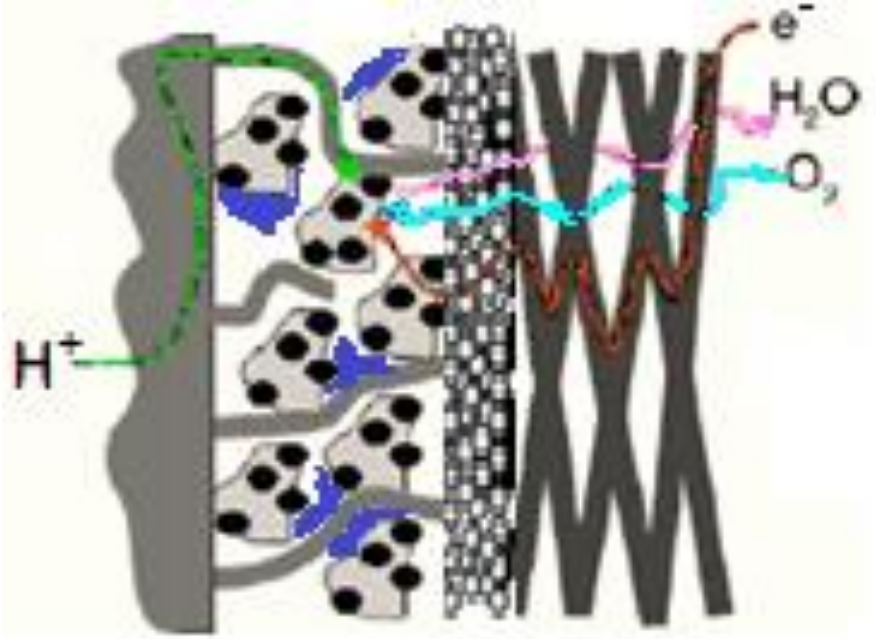
without CL microstructure

with CL microstructure

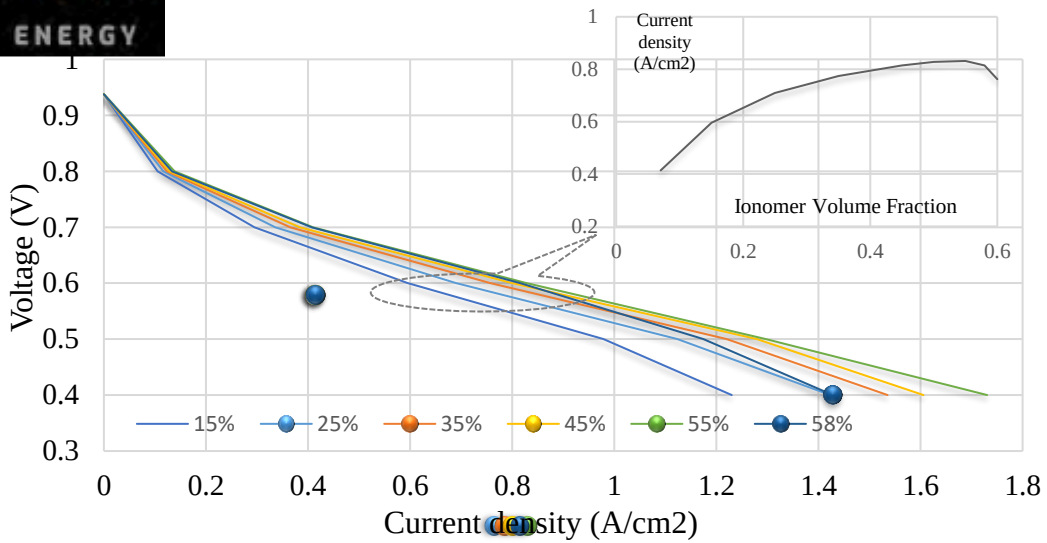
Water generation captured by the CPM model

Catalyst microstructure influence on PEMFC performance

- *The effect of ionomer volume fraction on current density*
- *The effect of Pt loading on current density*
- *The effect of catalyst agglomerate radius on current density*



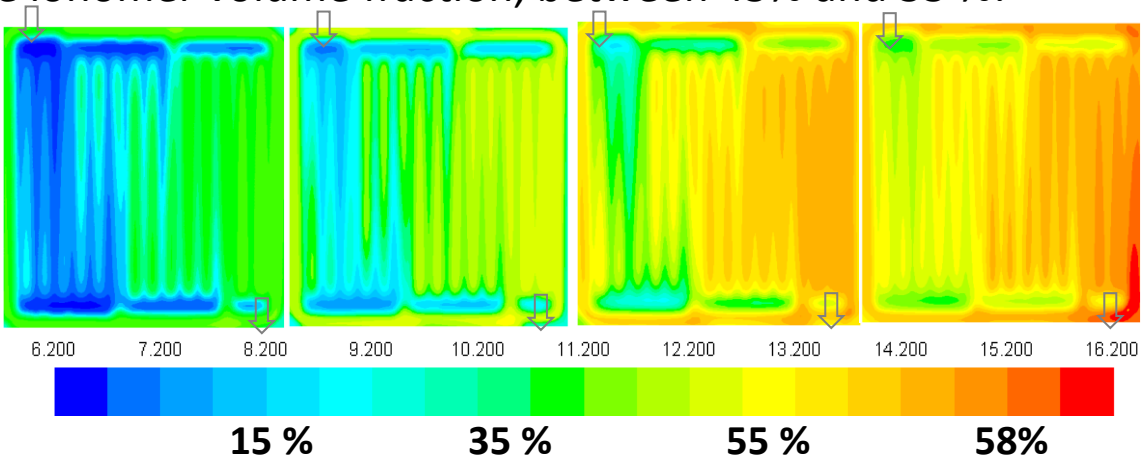
Ionomer volume fraction influence on PEMFC performance



The ionomer has two roles: for proton transfer and for binding the catalyst electrodes.

- there is an optimum range for the ionomer volume fraction, between 45% and 55 %.

Water content profile



-**vapor phase**: complete hydration of the ionomer takes place when the max. value of the water content is 14

-**liquid phase**: the max. value of the water content is 22, in the range 14-22 the flooding phenomenon occurs

Pt loading influence (m_{Pt}) on PEM fuel cell performance

The catalyst layer porosity is determined taking into account its composition: platinum dispersed carbon, ionomer and void:

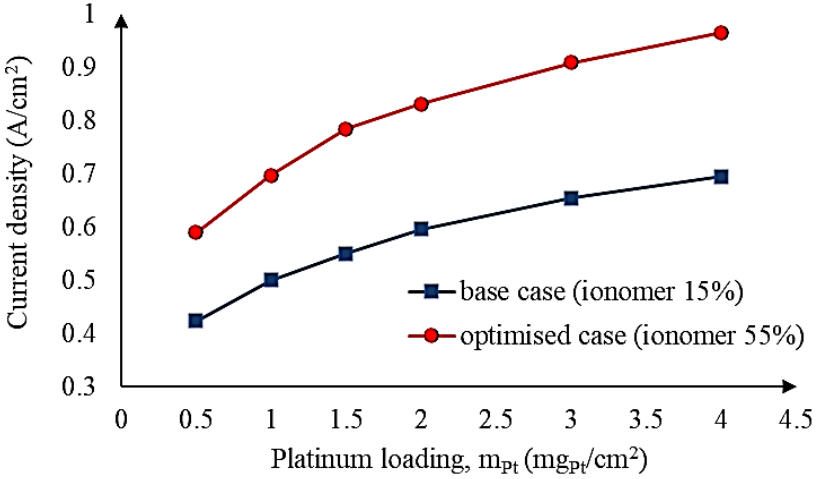
$$\varepsilon_{CL} = 1 - \varepsilon_{N,CL} - L_{GDL,CL} \cdot (1 - \varepsilon_{GDL}) - \varepsilon_{Pt} - \varepsilon_C$$

$$\varepsilon_{Pt} = m_{Pt}/(\delta_{CL} \cdot \rho_{Pt})$$

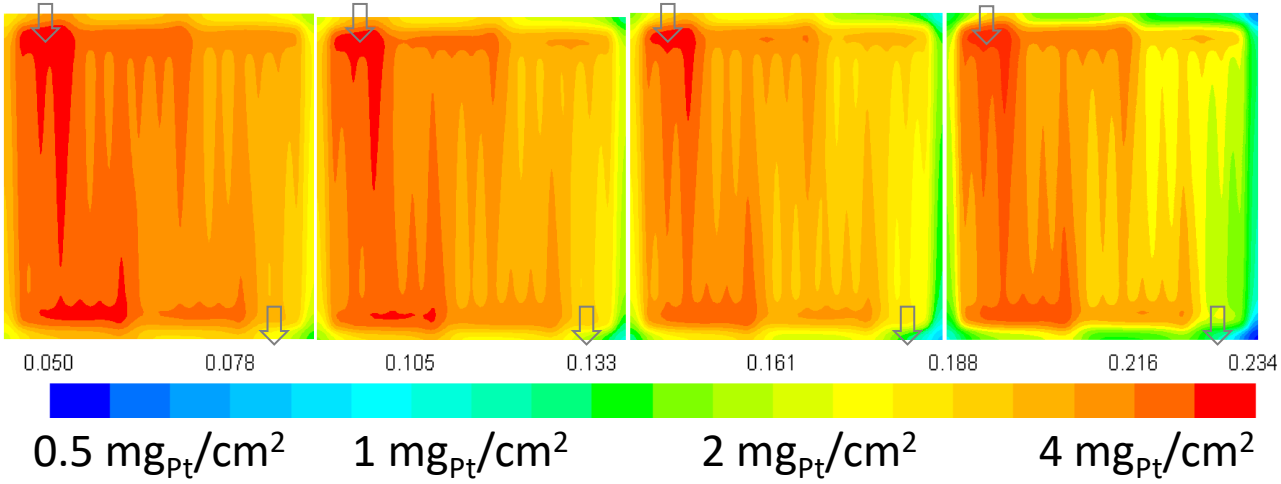
$$\varepsilon_C = m_{Pt} \cdot (1 - f)/(f \cdot \delta_{CL} \cdot \rho_C)$$

$$f = m_{Pt}/(m_{Pt} + m_C)$$

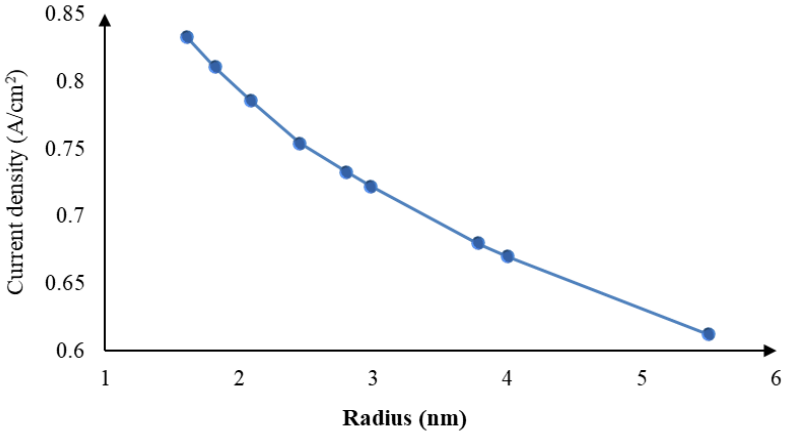
- the current trend in developing catalyst layers for PEM fuel cells is to reduce the Pt loading to 0.125 mg_{Pt}/cm^2 or to lower values
- A fast reaction rate for the oxygen reduction reaction takes place using a high platinum loading and this is due to the additional catalytic surface area provided



Oxygen mass fraction profile at 0.6V

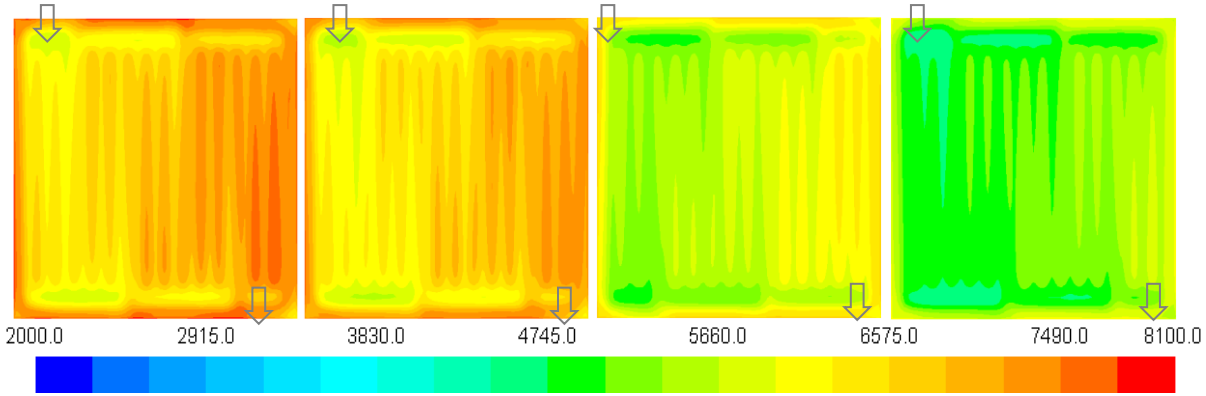


Pt particle radius influence on PEM fuel cell performance



- As the agglomerate radius increases the performance of the fuel cell deteriorates → smaller agglomerates pose smaller resistance to mass transport

Current density profile



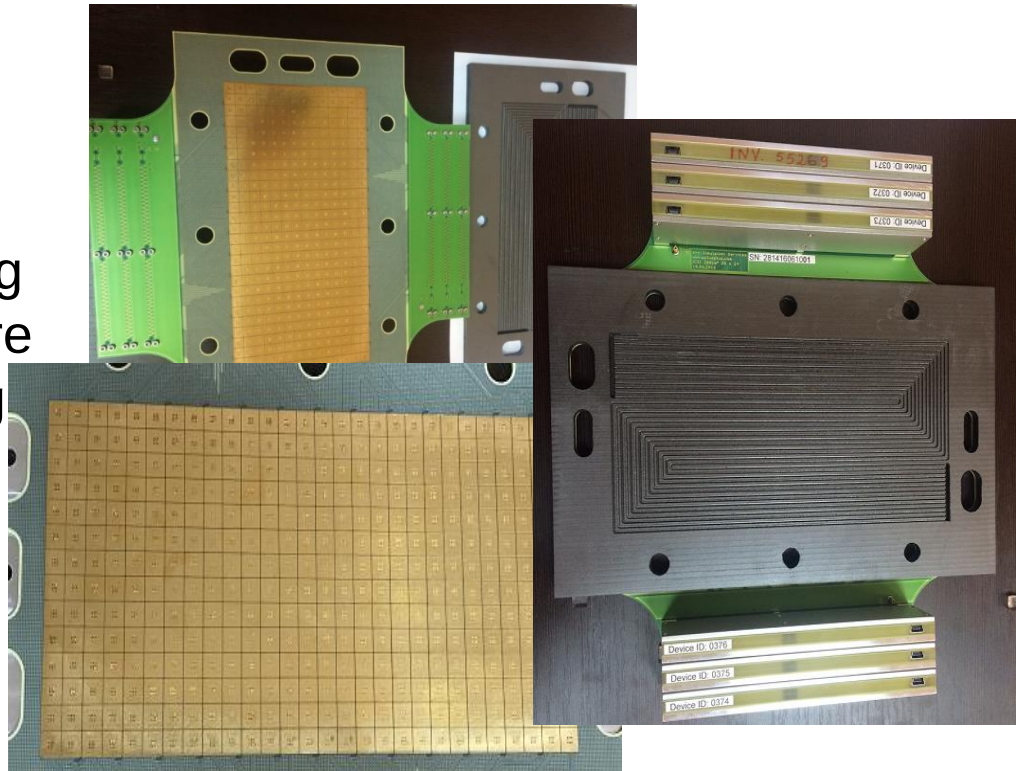
1.83nm **2.45 nm** **3.78 nm** **5.5 nm**

- a range between 2 and 5 nm for obtaining a maximum ORR mass activity and a better performance
- around 25% loss in the performance takes place when the radius of the platinum particles is increases from 1.61 nm to 5.5 nm.

CONCLUSIONS AND FUTURE WORK

- CFD investigations on PEM fuel cells based on ANSYS Fluent and Fuel Cell Module can be carried out in order to evaluate the effect of some design parameters on the performance.
- The CFD results must be validated against experiments.
- The design parameters must not be considered alone – operating parameters and materials properties must be considered together in a performance analysis.

- Integration of a system for mapping the current density and temperature for model validation and for getting information regarding the phenomena that are taking place inside each segment (14 x 28) of the system.



Thank you for your attention!