PEGASUS

PEMFC based on platinum group metal free structured cathodes

PEGASUS
PGM Free Catalyst

European Hydrogen Week

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#PRD2022
#CleanHydrogen

Project Overview

- Call year: 2017
- Call topic: FCH-01-2-2017 - Towards next generation of PEMFC: Non-PGM catalystsProject
- dates: 01 February 2018 - 30 June 2021
- % stage of implementation 01/11/2019: 100%
- Total project budget: 2,7 M€
- Clean Hydrogen Partnership max. contribution: 2,7 €
- Other financial contribution: 0€
- Partners: cea, DLR Deutsches Zentrum für Luft- und Raumfahrt German Aerospace Center, TUM, CSIC Consejo Superior de Investigaciones Científicas, ARMINES, Heraeus, IRD Fuel Cells, TOYOTA

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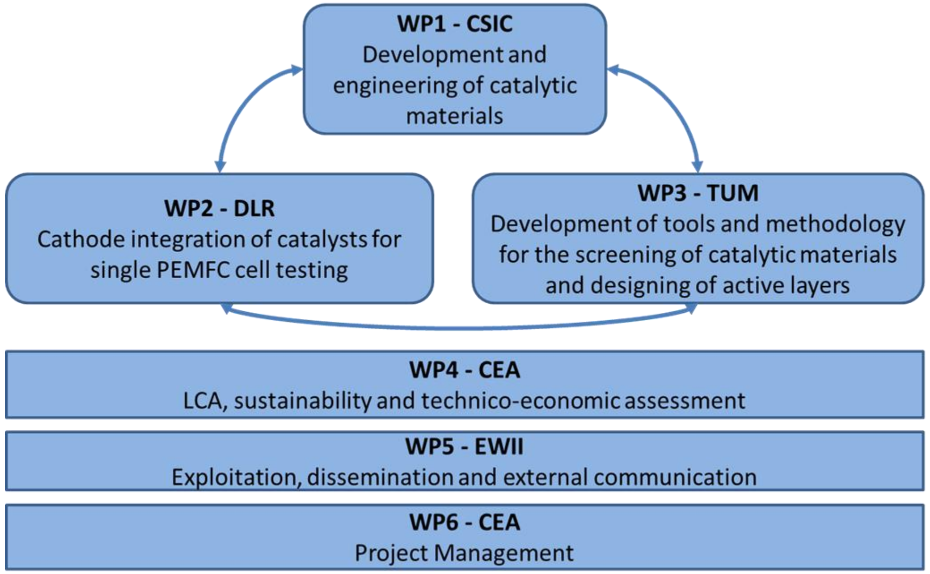
PEGASUS : Objectives

- Develop Platinum Metal Group (PGM) free catalysts for the cathode side of PEMFC
  - Only PEMFC is addressed (acidic conditions)
  - Only cathode side is addressed (Pt loading is ~4 times higher at cathode than anode)
  - 3 main levers :
    - ➢ Active site – intrinsic activity / active site density.
    - ➢ Catalyst structure – active site accessibility
    - ➢ Active layer structure – active site accessibility
- Manufacturability / processability
  - Process cost / LCA and upscale
  - PMF (Precious Metal Free) and CRM (Critical Raw Material) free
  - Environment friendly raw material.



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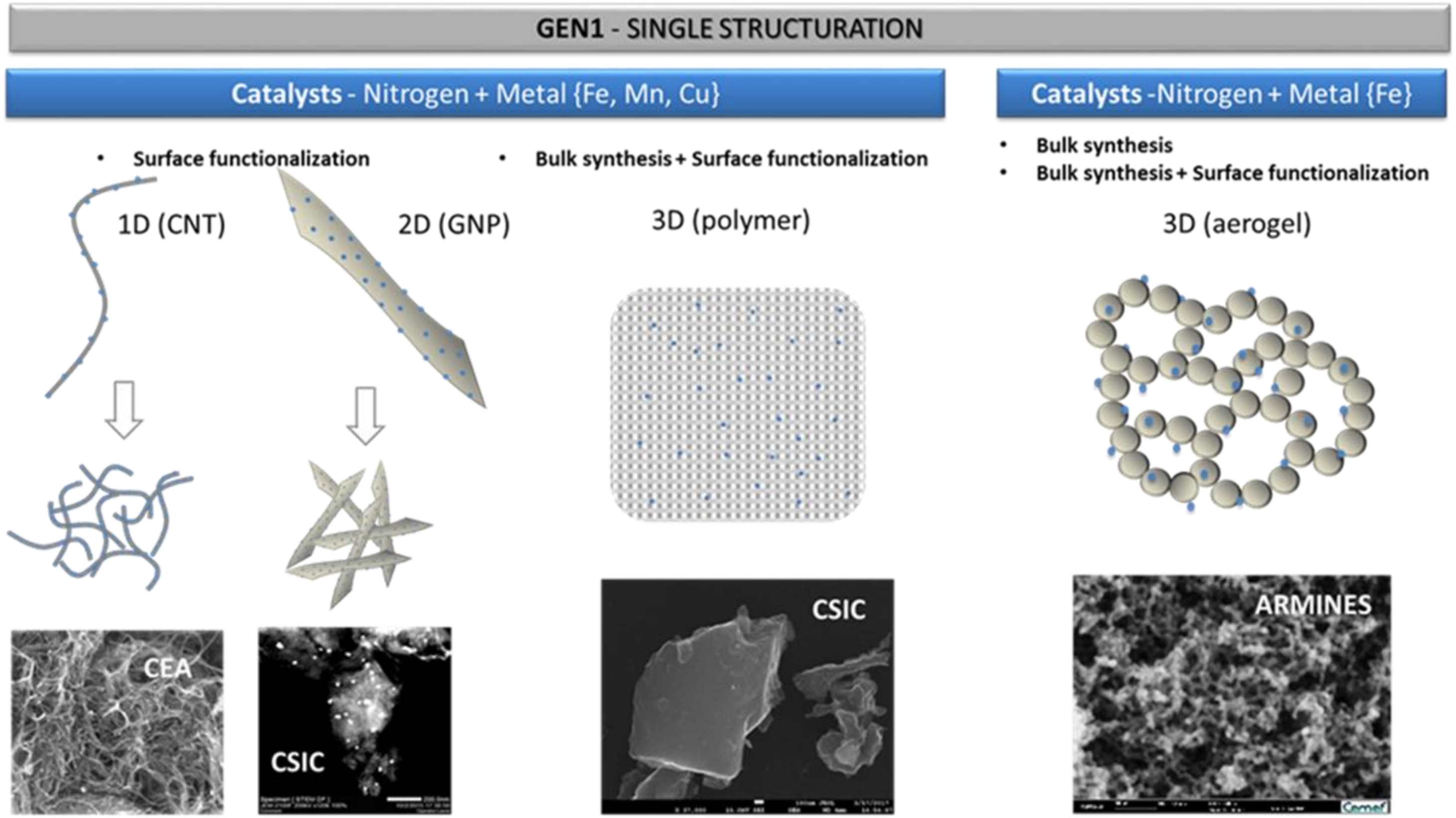
Project: Concept

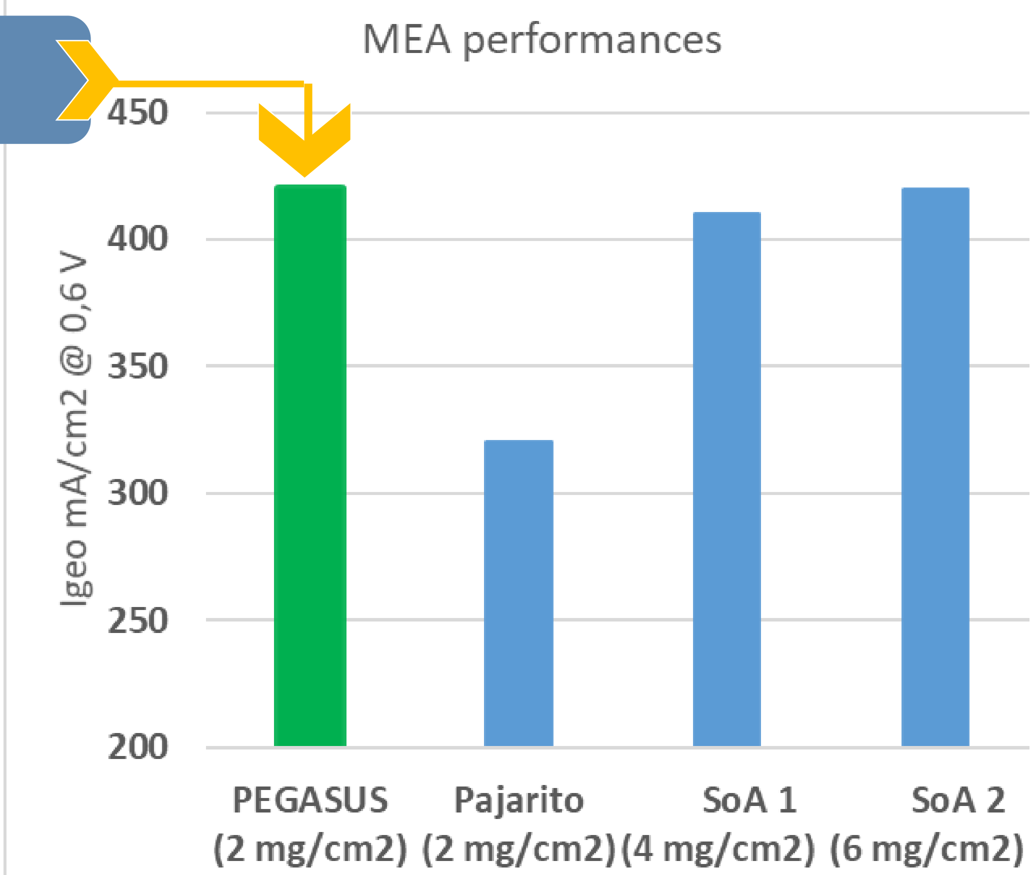


Progress: catalyst activity / MEA performances

600 mA @ 0,7 V
(420 mW)

25% 50% 75%

PEGASUS Best 420 mA @ 0,6 V
(250 mW)



1,5 bara / high flow cell (H2/Air) / 80°C/ 100%HR

- PEGASUS catalysts:
  - More than 65 g PGM free cat delivered (4 cat. technologies)
  - Characterization in RDE and MEA (1-5-25 and 50 cm^2 cells)
  - Impact of MEA fabrication process studied

- For the most active catalyst :
- Activity : 4.7 $^{+}/_{-}$ 0.5 A/g @ 0.80 V/RHE (HClO4; 25°C, O2 std, 1600 RPM)
- Cathode cat loading: 2 mg_{cat}/cm^2 (2 times less than SoA)
  - MEA: 5 times less power than Pt based MEA
  - The gap PGM free / Pt is higher with larger cell

SoA 1: Jiao, L. et al, Nat. Mater. (2021). https://doi.org/10.1038/s41563-021-01030-2
SoA 2: Mengjie Chen et al, 2021, J. Electrochem. Soc. 168, 044501

Progress: Deep charaterisation

Active site quantification / Active layer modelling / Operando charac.

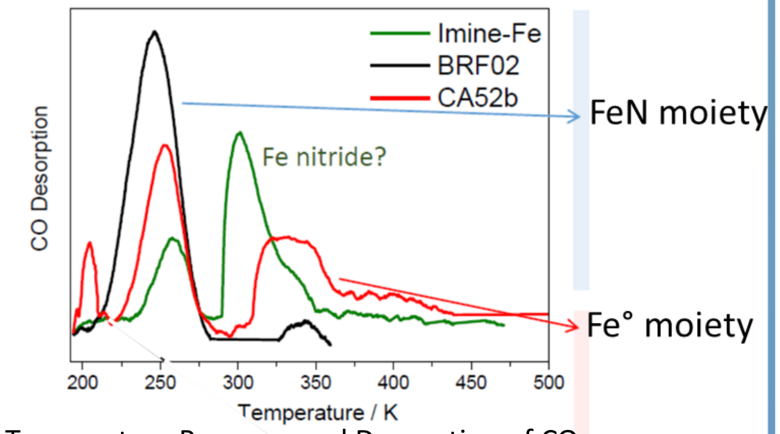
SECM Charac. / AFM /Neutron imag.
Active site quantification

25% 50% 75%

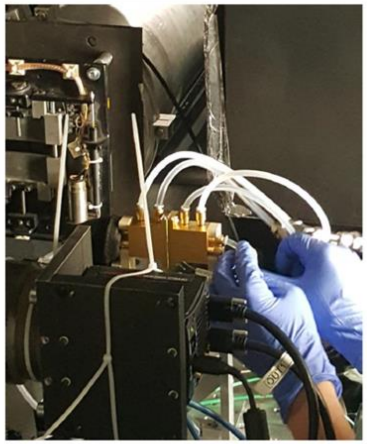
- Quantification of active site (TPD-CO) : 2.4 . 10 [20] sit/g_{cat} (BRFO2 – PEGASUS Best)
- ~ 10 times less than for Pt/C catalyst
- Only FeN moiety (acid washing not needed) / EXAFS / XANES
- NO – pulse chemisorption → 4 times active sites than measured made with TPD-CO

- Neutron Imaging – localisation of water inside the PGM free cathode MEA
  - H^+ transport is a bottleneck / cathode are too thick
  - Use of Low EW ionomer is beneficial

- Active layer modelling :
  - → densification of the active layer
  - → increasing the number of active sites /g_{cat}
  - → decreasing the radii of the agglomerate in the active layer



Temperature Programmed Desorption of CO


Installation of cell in set up for Neutron imaging

Progress - LCA and tech-eco assement

Performing Life Cycle Assessment
Tech-eco assessment

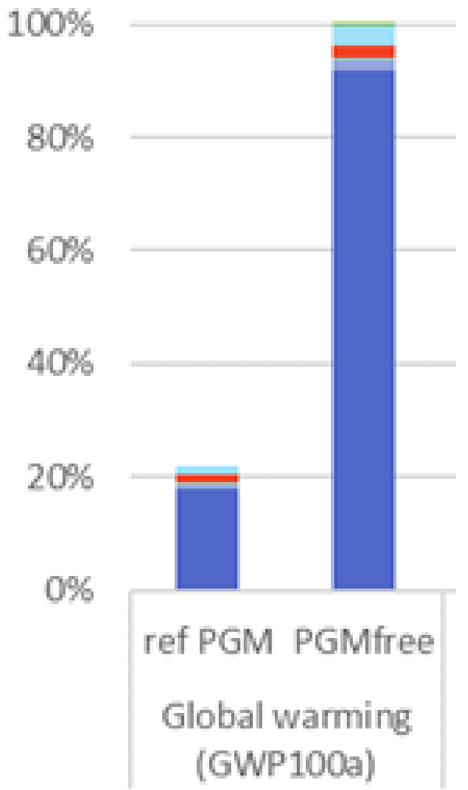
25% 50% 75%

- Tech-eco assessment (based on MEA Performance / at stack level only):
  - PEGASUS best cat. does not offer advantages Vs ref PGM[1] (stack cost + 300%)
  - New targets were defined :

  @ 780 mW/cm^2 for automotive in large area cell,
  the PGM free cat. start to be advantageous

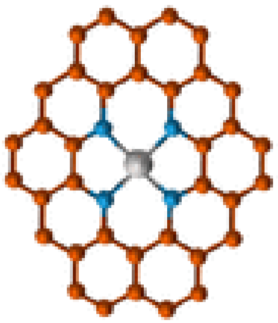
- Life cycle assessment (based on MEA Performance / at stack level only[1])
  - PEGASUS best cat. does not offer advantage Vs ref PGM[1] (global warming : x5)
  - This is mainly due to higher amount of material need (ionomer, GDL)
  - If the PEGASUS best MEA gave the same performance as Pt based MEA

  The save in global warming would be 10%



1: reference of the Pt based stack (ref PGM) : public data from Autostack CORE

Risks, Challenges and Lessons Learned



➤ Challenges:

- The active sites density for PGM free catalyst need to be improved by a factor 5 -10
- From geometrical point of view ,regarding the size of the FeN moiety, this not seems to be possible.
- For PEMFC (acidic condition), new PGM free material based on new active site structure should be investigated
- Those new material should be robust in operating PEMFC conditions

→ The thickness of the active layer needs to be 10 µm

→ Limit the H^+ transport loss

→ Use less PFSA based ionomer

→ Reduce the cost of the MEA and having a more beneficial LCA

Exploitation Plan/Expected Impact

Exploitation

- PGM free catalysts developed in PEGASUS are still at low TRL but :

- Use PEGASUS outcomes to design and fabricate AL for conventional MEA with increased performance.
- Use new and innovative developed carbon as support or catalyst in AEMFC
- Use developed LCA for other PEMFC catalysts

➡ Pegasus final WS was organized in June 2021.

Impact

-Giving a clear vision of the capabilities pf Fe/N/C PGM free catalyst for PEMFC application.

-Better understanding of the limitations / bottlenecks and challenges for the use of PGM free catalysts for PEMFC and thick MEA

-Creation of knowledge for the use of new carbon structure as support / active layer design for conventional MEA.

➡ 9 open access publications in Peer-reviewed jounals

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MANY THANKS

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Publications : part #1

Y. Wang, One-pot synthesis of Fe-N-containing carbon aerogel for oxygen reduction reaction, Electrocatalysis, 12(1), 78-90 2021 DOI: 10.1007/s12678-020-00633-8
GREEN open access : https://hal-mines-paristech.archives-ouvertes.fr/hal-03080371/document

P. Toudret, Impact of the Cathode Layer Printing Process on the Performance of MEA Integrating PGM Free Catalyst, *Catalysts* **2021**, *11*(6), 669; **https://doi.org/10.3390/catal11060669**
GOLD open access : https://www.mdpi.com/2073-4344/11/6/669/htm

O. Henrotte, Steady-State Electrocatalytic Activity Evaluation with the Redox Competition Mode of Scanning Electrochemical Microscopy: A Gold Probe and a Boron-Doped Diamond Substrate; ChemElectroChem 2020, 7, 4633 –4640, https://doi.org/10.1002/celc.202001088
GREEN Open access : https://hal-cea.archives-ouvertes.fr/cea-03014550/document

N. Limani, Local probe investigation of electrocatalytic activity, Chem. Sci., 2021, 12, 71; DOI: 10.1039/d0sc04319b
GOLD open access : https://pubs.rsc.org/en/content/articlelanding/2021/SC/D0SC04319B

Á. García, Study of the evolution of FeN*x*C*y* and Fe3C species in Fe/N/C catalysts during the oxygen reduction reaction in acid and alkaline electrolyte, J. Power Sources , Volume 490, 2021, 229487; https://doi.org/10.1016/j.jpowsour.2021.229487
GOLD open access : https://www.sciencedirect.com/science/article/pii/S0378775321000380?via%3Dihub

Á. García, Fe doped porous triazine as efficient electrocatalysts for the oxygen reduction reaction in acid electrolyte, Applied Catalysis B: Environmental, Volume 264, 2020, 118507, https://doi.org/10.1016/j.apcatb.2019.118507
GOLD open access : https://www.sciencedirect.com/science/article/pii/S0926337319312536?via%3Dihub

Publications : part #2

D.Garcia-Sanchez, Local impact of load cycling on degradation in polymer electrolyte fuel cells, Applied Energy Volume 259, 2020, 114210, https://doi.org/10.1016/j.apenergy.2019.114210
GOLD open access : https://www.sciencedirect.com/science/article/pii/S0306261919318975?via%3Dihub

Y. Wang,, Influence of the synthesis parameters on the Proton Exchange Membrane Fuel Cells performance of Fe-N-C aerogel catalysts, Journal of Power Sources, Volume 514, 2021, 230561, https://doi.org/10.1016/j.jpowsour.2021.230561
GREEN open access: https://hal.archives-ouvertes.fr/hal-03361259/document

A. García, Effect of the thermal treatment of Fe/N/C catalysts for the oxygen reduction reaction synthesized by pyrolysis of covalent organic frameworks, Ind. Eng. Chem. Res. 2021, 60, 51, 18759–18769, https://doi.org/10.1021/acs.iecr.1c02841
GOLD open access : https://pubs.acs.org/doi/10.1021/acs.iecr.1c02841