Advanced Multi-Fuel **Re**former **for** Fuel **CELL** CHP Systems

ReforCELL

Newsletter – Issue 5 – 2nd half 2014

Editorial

Welcome to the new release of the ReforCELL newsletter. Since the previous newsletter different progress has been achieved. The fluidized bed reactor for autothermal reforming has been proven at lab scale. The results identified difficulties in maintaining good sealing conditions for continuous operation at elevated temperatures under reactive condition. Modifications to the sealing joints have been defined and will be implemented for the pilot reactor. In addition, the design of the pilot reactor has been achieved and is now ready for construction. Latest news on the activities developed in the project can be found in this newsletter.

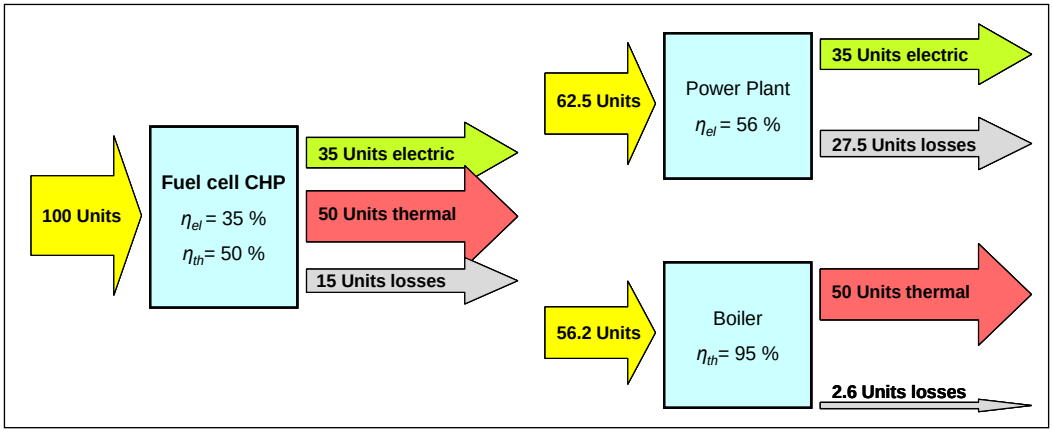
Dissemination and collaboration with other major European Projects are a major point for the ReforCELL project. More than 19 publications and/or conference participations have been presented by the ReforCELL consortium. Besides, a joint workshop with other EU projects (CARENA, DEMCAMER and CoMETHy) is scheduled in November 2014 in order to share experiences in the field of membranes and membrane reactors. Reforcell partners will give presentations on both membrane development and reactor testing (at lab scale) and design (at larger scale).

In this Issue:

What is ReforCELL?

Concept

Stationary fuel cells offer a clean and efficient source of electricity in systems ranging from 1 kW up to 1 MW or more. With appropriate fuel processing technology, fuel cells are able to tap into established or accessible sources of fuels such as natural gas but also various other fuels including biofuels and bio-gases. With cogeneration or combined heat and power (CHP), efficiencies improve dramatically from 30–50 % up to as high as 80-90 % with significant primary energy savings. In spite of these demonstrated benefits, cost and reliability issues make the technologies long-term potential difficult to predict. In order to reduce costs and increase the reliability of the technology, work must be done on fuel processing design and system optimization.



Fuel cell based Combined Heat and Power

ReforCELL aims at developing a high efficiency CHP system based on:

i) Design, construction and testing of an advanced catalytic membrane reactor for pure hydrogen production from reforming and optimization of the reactor components (catalyst, membranes, heat management etc.)

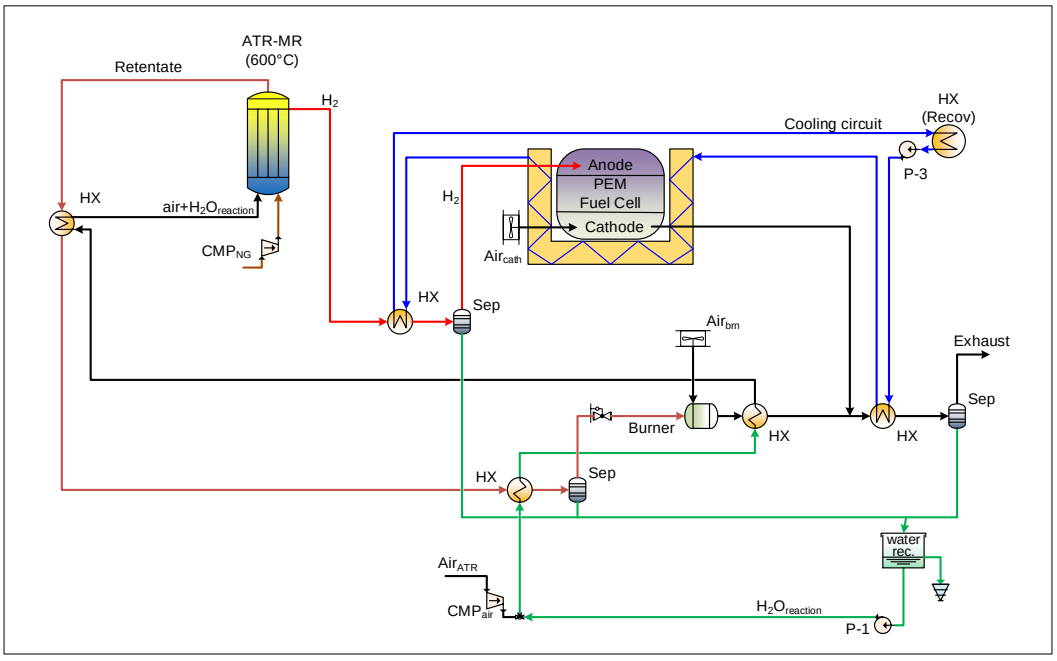
ii) Design and optimization of all the components of CHP system for the integration of the membrane reformer to the fuel cell stack.

The main idea of ReforCELL is to develop a novel more efficient and cheaper membrane reactor in order to intensify the process of hydrogen production through the integration of reforming and purification in one single unit.

Project objectives

ReforCELL aims at developing a high efficiency (above 42 % electric and 90 % total) PEM fuel cell micro Combined Heat and Power system based on a novel, more efficient and cheaper pure hydrogen production unit, together with optimized design of the subcomponent for the BoP.

The main objective of ReforCELL is to develop a new catalytic membrane reactor for pure hydrogen production (5 Nm^3/h) from hydrocarbon reforming in order to intensify the process of hydrogen production. The target will be pursued with the integration of the reforming and purification in one single unit. The novel reactor will be more efficient than the state-of-the-art technology due to an optimal design aimed at circumventing mass and heat transfer resistances. Moreover, the design and optimization of the subcomponents for the BoP will be also addressed.



System schematic layout

This objective is directly related to the development of a novel catalytic membrane reactor (CMR) for hydrogen production with:

- Improved performance (high conversion at low temperature for the autothermal reforming reaction);
- Enhanced efficiency (reduction of the energy consumption);
- Long durability under CHP system working conditions.

Scientific and Technical challenges

The technical objectives needed to achieve these goals with the novel catalytic membrane reactor are:

- Develop an advanced catalyst suitable for different reforming reactions under moderate (<700°C) conditions and resistant to sulphur compounds and coke formation and at reduced cost;
- Develop new hydrogen permeable membrane materials with improved separation properties, long durability, and with reduced cost, to be used under reactive conditions;
- Assess the large scale production of the membrane developed;
- Understand the fundamental physico-chemical mechanisms and the relationship between structure/property/performance and manufacturing process in membranes and catalysts;
- Design, model and build a novel more efficient (e.g. reducing the number of steps) multi-fuel catalytic membrane reactor configurations for small-scale pure hydrogen production;
- Validate the new membrane reactor configurations, and design a semi-industrial Autothermal Reforming (ATR) prototype for pure hydrogen production;
- Improve the cost efficiency of membrane reactors by increasing their performance, decreasing the raw materials consumption and the associated energy losses.

Other technical objectives are related to the integration and validation of the multi-fuel reformer into the PEM fuel cell CHP system:

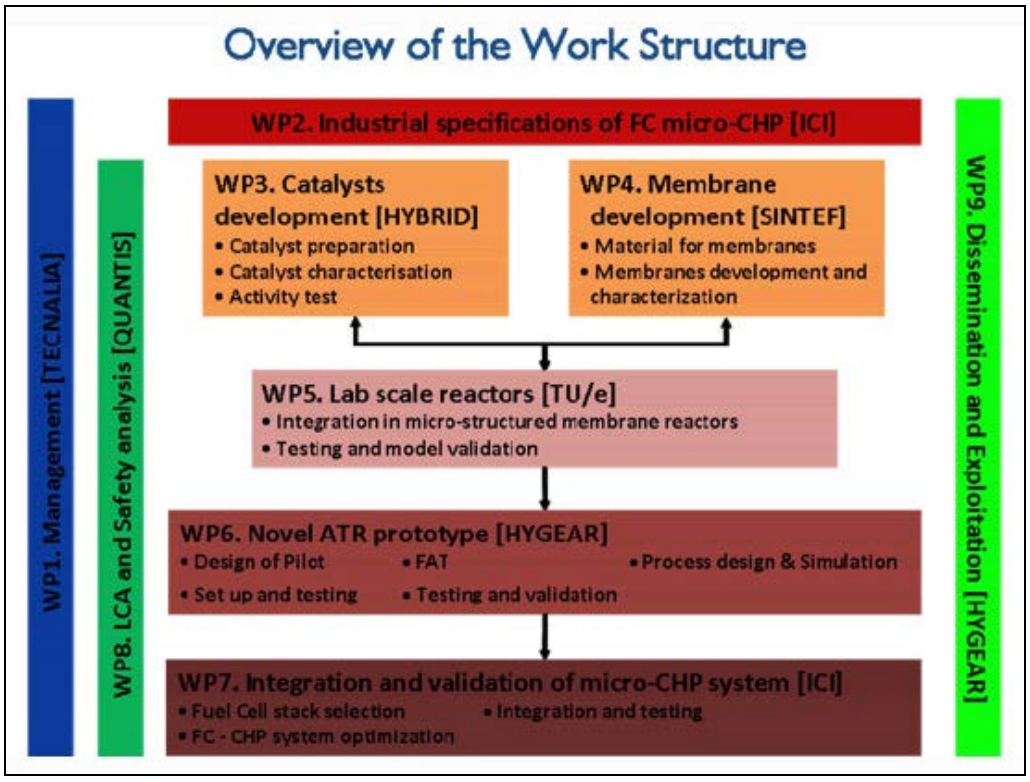
- Design the optimum CHP system (aided by simulation tools);
- Test the reliability of the novel reactor with a Fuel Cell CHP system;
- Assess the health, safety and environmental impact of the system developed, including a complete Life Cycle Assessment (LCA), of the developed system.

Partnership

For a maximum impact on the European industry, ReforCELL is carried out by a multidisciplinary and complementary team consisting of 11 top level European organisations from 6 countries: 6 Research Institutes and Universities and 5 industrial partners from different sectors (from hydrogen producer and catalyst developer to boilers and systems integrators). In this way, the ReforCELL brings together solid experience in conception and erection of hydrogen production systems, catalyst and membrane development, system integration, plant optimization, LCA and industrial risk analyses.

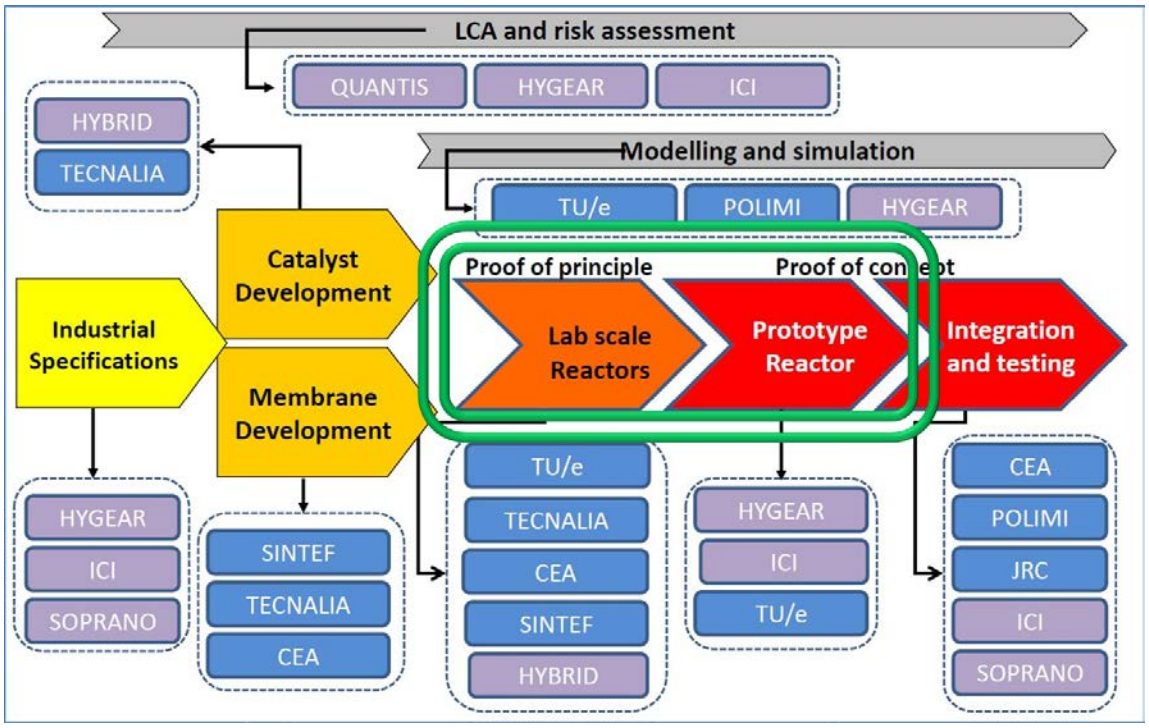
Project structure

The project is structured and divided into nine well-defined objective-oriented work packages following the focus on efficiency improvement of the overall m-CHP system based on PEM fuel cell and innovative multi-fuel processor.



ReforCELL in progress

ReforCELL project has been ongoing with important results in the lab scale prototype as well as reactor design. The fluidized bed reactor for autothermal reforming has been proven at lab scale. Tests on the integration of novel catalyst made by HYBRID and membrane made by TECNALIA under fluidized bed were carried out. The results identified difficulties in maintaining good sealing conditions for continuous operation at elevated temperatures. The design of the pilot reactor has been achieved and is now ready for construction. The reactor will be first tested at HyGear and then integrated into the CHP system at SOPRANO facilities.



Novel catalysts and membranes for Autothermal Reforming - Membrane Reactor (ATR-MR)

Hybrid Catalysis has developed a methane reforming catalyst with outstanding performance. Several kilograms have been supplied to network partners. The catalyst outperforms commercially available systems with respect to stable operation at relatively low temperature and very high resistance to coke formation. This desirable performance originates in the application of a novel redox active support. Catalyst material is available in several sieve fractions together with inert filler having the same bulk density. These features make it suitable for virtually any reactor concept.

SINTEF has developed a micro-channel configured membrane module with channel dimensions between 200 and 1000 micron. Introduction of a porous permeate section increased the pressure tolerance to more than 15 bar feed pressure. Intermetal diffusion barrier layers are currently being implemented to operate at temperatures above 450 °C. An enlarged micro-channel module is also being developed. SINTEF has also prepared tubular-supported membranes with very good selectivities achieving ReforCELL criteria for operation temperature relevant to the reforming of ethanol. In addition, SINTEF has also performed long term temperature and stability testing of membranes produced at TECNALIA.

TECNALIA has developed thin Pd-Ag membranes supported on ceramic tubes that show high hydrogen permeance and selectivity achieving the ReforCELL project targets, and good

long-term stability at 600ºC (operating temperature for ATR-MR). The activities on the last months have been focused on developing Pd-Ag membranes supported on metallic tubular supports with intermetallic diffusion barrier layer to be also integrated in the fluidized bed ATR-MR. Long term temperature and stability testing carried out at TU/e show that high selectivity can be obtained (H_2/N_2> 200,000) when testing between 500 °C – 600 °C but after 800 hours N2 leakage was detected through the membrane (H_2/N_2 = 2,650). Work on Pd-Ag membranes supported on metallic tubular supports is still on-going.

Membrane reactor development

The fluidized bed reactor for autothermal reforming has been proven at lab scale. The catalyst was operated continuously for more than 4 weeks while the reactor was successfully tested with commercial membranes. Tests on the integration of novel catalyst and membrane under fluidized bed were also carried out. The results identified difficulties in maintaining good sealing conditions for continuous operation at elevated temperatures. Modifications to the sealing joints have been defined and are implemented for the pilot reactor.

Industrial specifications, layout and operation of CHP

The design of the pilot reactor has been achieved and is now ready for construction. The reactor will be first tested at HyGear facilities in Arnhem, The Netherlands and then integrated into the CHP system at SOPRANO facilities in Vaulx-Milieu, France.

Life cycle assessment

The screening life cycle assessment (LCA) has been performed. The data used in input to calculate the first draft results for a standard PEM micro-CHP scenario will need to be refined through a second data collection/validation step with the project partners according to the iterative nature of LCA. For the next step, data will also be collected and refined to establish the ReforCELL developed scenario.

Highlights

Workshop on Scale-up of Pd Membrane Technology: From Fundamental Understanding to Pilot Demonstration (*20&21 November 2014, ECN, Petten, The Netherlands)*

Thanks to their outstanding hydrogen selectivity, palladium membranes have attracted extensive R&D interest in the 21st century with promising "breakthrough" applications for hydrogen power, refining and petrochemicals, hydrogen vehicles and many more. The workshop is the follow-up of the first "Pd-membrane Scale-Up" workshop (Roma, Italy, November 2012), a unique knowledge-sharing experience for both the EU-funded organizing projects and all participants. This experience has strengthened the believe that there is a need and a ground for a second workshop, where the whole Pd-membrane R&D spectrum "From Fundamental Understanding to Pilot Demonstration" will be further

explored together by representatives of academia, research institutions and industrial stakeholders.

The workshop is organised by 4 projects funded by the EU through FP7: CARENA & DEMACAMER under the NMP priority and CoMETHy & ReforCELL under the FCH-JU priority. All the projects have commonality and synergy in their research objectives. The 2-day program provides an overview about the critical topics for Pd membrane technology scale-up:

- Fundamentals of Pd membranes
- Support & Seals manifacturing
- Membrane module design & system integration
- Lab-scale long-term stability testing results
- Industrial power plant operational insights

REGISTRATION:

Please the following link can be used for the registration:

https://www.ecn.nl/registration/PdMembraneWorkshop/

The deadline for registration is the 17th of October 2014. After registration a confirmation e-mail will be sent including information on accommodation and transport from hotel to workshop venue at ECN.

Dissemination activities, publications and presentations:

- L. Roses, S. Campanari, G. Manzolini, *Computational fluid dynamics (CFD) analysis of membrane reactors: simulation of a palladium-based membrane reactor in fuel cell micro-cogenerator system*, in book: A. Basile "Handbook of membrane reactors: Fundamental materials science, design and optimisation (Vol. 1). ISBN-13: 9780857094148. Woodhead Publ. 2013.

- G. Manzolini, D. Jansen, *Economic analysis of systems for electrical energy and hydrogen production: fundamentals and application to two membrane reactor processes*, in book: A. Basile "Handbook of membrane reactors: Reactor types and industrial applications (Vol. 2). ISBN-13: 9780857094155. Woodhead Publ. 2013.

- F. Gallucci, E. Fernandez, P. Corengia, M. van Sint Annaland, *Recent advances on membranes and membrane reactors for hydrogen production*, Chem. Eng. Sci. 92 (2013) pp.40-66.

- Peters, T.A., Sunding, M.F., Stange, M., Bredesen, R., *Stability investigation of micro-configured Pd-Ag membrane modules – effect of operating temperature and pressure*, Int. J. Hydrogen. Energy, (2014) submitted.

- Peters, T.A., Kaleta, T., Stange, M., Bredesen, R., *Pd-based Membranes in H2 Production and CO2 Capture Processes: status at SINTEF*, in Proc. IMeTI & CARENA Workshop. (March 27-28, 2012), Editors: -, Montpellier, France, (2012).

- L. Marra, F. Gallucci, T.A. Peters, M. Stange, R. Bredesen, M. van Sint Annaland, *Methane ATR in a catalytic microreactor and integration of a self-supported Pd-membrane for hydrogen separation*, 9th European Congress of Chemical Engineering (ECCE), 21-25 April 2013. The Hague, The Netherlands

- L. Marra, F. Gallucci, T.A. Peters, M. Stange, R. Bredesen, M. van Sint Annaland, *Auto-Thermal Reforming over RhZrO2 catalyst in a membrane Microreactor*, 11th International Conference of Catalysis in Membrane Reactors (ICCMR11), 7-11 July 2013, Porto, Portugal

- T.A. Peters, M. Stange, R. Bredesen, *Microchannel-supported thin Pd-alloy membranes – application in membrane micro-reactors for methane steam reforming and propane dehydrogenation processes*, 11th International Conference of Catalysis in Membrane Reactors (ICCMR11), 7-11 July 2013, Porto, Portugal

- D.A. Pacheco Tanaka, *Preparation and characterization of Palladium membranes for hydrogen separation*, 7-11 July 2013, Inorganic membranes summer school, Valencia, Spain.

- L. Marra, P.F. Wolbers, F. Gallucci and M. van Sint Annaland, *Development of a RhZrO2 catalyst for low temperature autothermal reforming of methane in membrane reactors*, Accepted Catal. Today - http://dx.doi.org/10.1016/j.cattod.2013.10.069

- J.L. Viviente, *Design and manufacturing of catalytic membranes reactors (DEMCAMER) and advanced multi-fuel reformer for CHP-fuel cell systems (REFORCELL)*, Inorganic membranes summer school, 4-6 September 2013, Valencia, Spain.

- L. Marra, F. Gallucci, T.A. Peters, M. Stange, R. Bredesen, M. van Sint Annaland, *Membrane microreactors for hydrogen production via ATR of methane*, 4th International Conference on Structured Catalysts and Reactors, 25-27 September 2013, Beijing, China.

- D.A. Pacheco Tanaka, *Reparation of supported Pd based membranes for hydrogen separation*, 5th World hydrogen Technologies Convention, 25-28 September 2013, Shangai, China.

- *ReforCELL public poster presentation,* Dissemination of European Projects section, 5th European Fuel Cell Conference, 11-13 December 2013, Rome, Italy.

- D.A. Pacheco Tanaka, *Palladium membranes for hydrogen separation and membrane reactors*, IX Congreso Ibero-Americano en Ciencia y Tecnología de Membranas (CITEM), Santander (Spain), 25-28 May, 2014

- D.A. Pacheco Tanaka, F. Gallucci, E. Fernandez, A. Helmi, J. Melendez, K. Coenen, M. Van Sint Annaland, *Fluidized bed membrane reactors for H2 production using thin Pd-Ag supported membranes*, 13th International conference on inorganic membranes, ICIM-13, July 6-9, Brisbane, Australia, (2014)

- E. Fernandez, J. Melendez, D.A. Pacheco Tanaka, L. Marra, I. Ottenheijm, M. van Sint Annaland, F. Gallucci, *Development of Pd-Ag supported membranes for high temperature fluidized bed membrane reactors,* 13th International conference on inorganic membranes, ICIM-13, July 6-9, Brisbane, Australia, (2014)

- Peters, T.A., Stange, M., Bredesen, R., *Fabrication of Pd-based membranes by magnetron sputtering - possibilities for membrane and module design*, in Palladium Membrane Technology Scale-up, Woodhead Publishing, 2014, submitted.

- Stange, M., Peters, T.A., Bredesen, R., *Stability studies of microchannel-supported thin Pd-alloy membranes,* in 13th International conference on inorganic membranes, ICIM-13, July 6-9, Brisbane, Australia, (2014).

Upcoming events

6 - 8 October 2014	14th Fuel cell forum for producers and users, Messe Stuttgart, Germany http://www.f-cell.de/english/service/outlook-2013/
12 - 13 November 2014	15th Aachener Membran Kolloquium, AMK Aachen, Germany http://www.avt.rwth-aachen.de/AMK/
20 - 21 November 2014	Scale-up of Pd Membrane Technology From Fundamental Understanding to Pilot Demonstration, ECN, Petten, The Netherlands http://www.reforcell.eu/pdfs_documentos/Workshop2014-flyer-final.pdf
9 - 12 December 2014	EMHYTEC, Euro-mediterranean Hydrogen technologies conference, Taormina, Italy http://www.itae.cnr.it/emhytec2014/
1 - 3 December 2014	CARISMA 2014 Conference, Cape Town, South Africa http://www.carisma2014.com/index.php
25 - 30 January 2015	9th Int. Symposium Hydrogen & Energy, EMMETTEN, Switzerland http://hesymposium.ch/
27 - 28 April 2015	Hydrogen + Fuel Cells Summit 2015, Vancouver, BC, Canada http://www.hfc2015.com/about/
22 - 25 June 2015	12th International Conference on Catalysis in Membrane Reactors (ICCMR12), Szczecin, Poland http://www.iccmr12.zut.edu.pl/index.php
28 June - 2 July 2015	ASME Power & Energy 2015, San Diego, CA http://www.asmeconferences.org/POWERENERGY2015/index.cfm
30 June - 3 July 2015	5th European PEFC & H2 Forum, KKL, Lucerne, Switzerland http://www.efcf.com/
6 - 10 September 2015	Euromembrane 2015, Aachen, Germany http://www.avt.rwth-aachen.de/Euromembrane2015/index.php
11 - 14 October 2015	6th WHTC World Hydrogen Technology Convention, Sydney, Australia http://www.whtc2015.com/
13 - 17 June 2016	21th WHEC World Hydrogen Energy Conference, Zaragoza, Spain http://www.whec2016.es/

ReforCELL in figures:

- *11 partners (4 SME; 1 IND; 6 RES)*
- *6 countries*
- *5.6 M€ project (2.9 M€ EU funded)*
- *Start February 2012*
- *Duration: 36 months*
- *Key milestones:*
  - *September 2013- Validation of lab-scale reactor*
  - *July 2014- Validation of the pilot scale prototype*
  - *January 2015- Validation of the m-CHP system*

Project manager:

Dr. José-Luis Viviente
TECNALIA
joseluis.viviente@tecnalia.com

Technical manager:

Dr. Fausto Gallucci
TU/e
f.gallucci@tue.nl

Dissemination manager:

Dr. Giampaolo Manzolini
Politecnico di Milano
giampaolo.manzolini@polimi.it

Exploitation manager:

Dr. Leonardo Roses
HyGear B.V.
leonardo.roses@hygear.nl

More information on ReforCELL (including a non-confidential presentation of the project) is available at the project website and FCH-JU website:

http://www.reforcell.eu

http://www.fch-ju.eu/project/advanced-multi-fuel-reformer-chp-fuel-cell-systems

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Disclosure:

The present document reflects only the author's views, and neither the FCH-JU nor the European Union is liable for any use that may be made of the information contained therein.

Advanced Multi-Fuel **Re**former **for** Fuel **CELL** CHP Systems

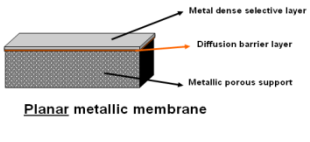
ReforCELL

Summary:

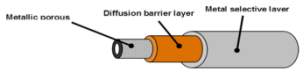
ReforCELL aims at developing a high efficiency (above 42 % electric and 90 % total) PEM based micro-CHP system through: i) design, construction and testing of an advanced catalytic membrane reactor for pure hydrogen production from hydrocarbon reforming, and ii) design and optimization of all the components for the integration of the membrane reformer to the fuel cell stack.

The main idea of ReforCELL is to develop a novel more efficient and cheaper membrane reactor in order to intensify the process of hydrogen production through the integration of reforming and purification in one single unit. In order to achieve this objective, novel stable catalysts and high permeable and more stable membranes need to be developed. Afterwards, suitable reactor design will be realized and tested at laboratory scale for later scaling up to prototype scale (5 Nm^3/h of pure hydrogen) and tested in a CHP system.

The connection of the novel fuel processor within the CHP will be optimized by designing heat exchangers and auxiliaries required in order to decrease the energy losses.

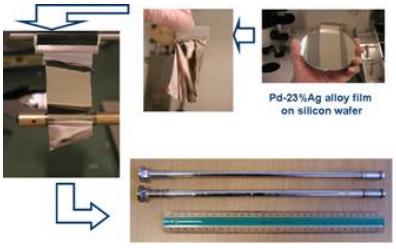


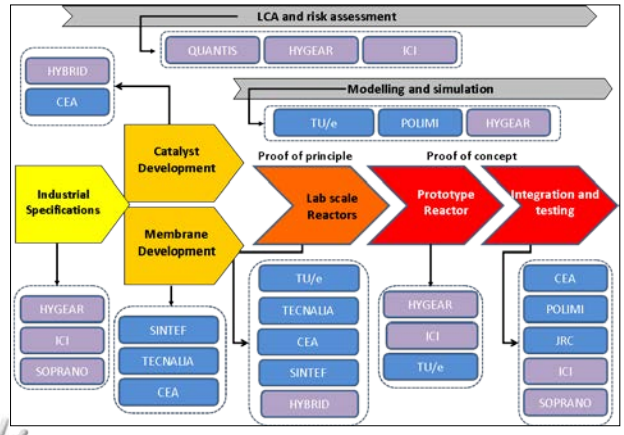
Planar metallic membrane

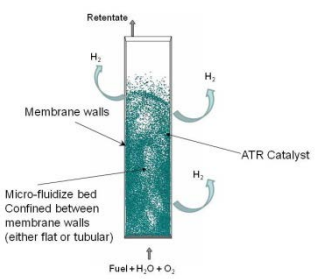


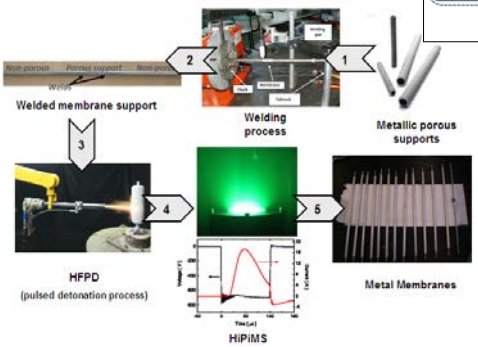
Tubular metallic membrane

Cross section SEM micrograph of Pd-based membrane









Contact:

TECNALIA
Materials for Energy - Energy Unit
Dr. José-Luis Viviente
Mikeletegi Pasealekua, 2
E-20009 Donostia - San Sebastián (Spain)
joseluis.viviente@tecnalia.com