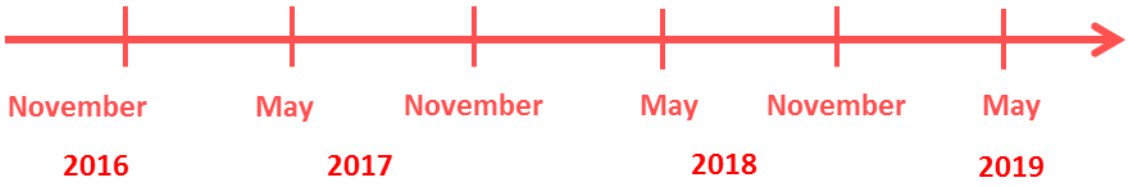
Flexible Hybrid separation system for H2 recovery from NG Grids

Newsletter – Issue 4 – December 2018



Editorial

Welcome to this fourth HyGrid newsletter. HyGrid is a three-year project targeting the development of a high performance, cost effective separation technology for the direct separation of pure hydrogen from natural gas grids. Three different technologies - membrane separation, electrochemical separation and temperature swing adsorption - will be combined in a new separation system to decrease the total cost of hydrogen recovery. The new separation & purification system will increase the value of hydrogen blended into the natural gas grid.

The present newsletter is the fourth release and it is presenting the progress on the project and highlighting information related to the R&D fields addressed. Hope you will find the info in this newsletter interesting. On our website www.hygrid-h2.eu you will find public presentations, all the public deliverables of the project and many other interesting news. Stay tuned!

In this Issue:

What is HyGrid?

The concept

One of the main problems for the implementation of the hydrogen-based economy is the transportation from production centres to the end user both industries and population. To solve this problem, besides the in-situ production of hydrogen, the use of the existing Natural Gas network has been proposed for storing and distributing hydrogen. However, cost effective separation technologies for direct separation of hydrogen from the natural network should be developed for separating and purifying the hydrogen to match the end user requirements.

The HyGrid project proposes an integral solution for developing of an advanced high performance, cost effective separation technology for direct separation of hydrogen from natural gas networks. By using a novel membrane based hybrid technology combining three technologies integrated in a way that enhances the strengths of each of them (Figure 1): membrane separation technology is employed for removing H_2 from the "low H_2 content" (e.g. 2-10 %) followed by electrochemical hydrogen separation (EHP) optimal for the "very low H_2 content" (e.g. <2 %) and finally temperature swing adsorption (TSA) technology to purify from humidity produced in both systems upstream, pure hydrogen production (ISO 14687) will be obtained.

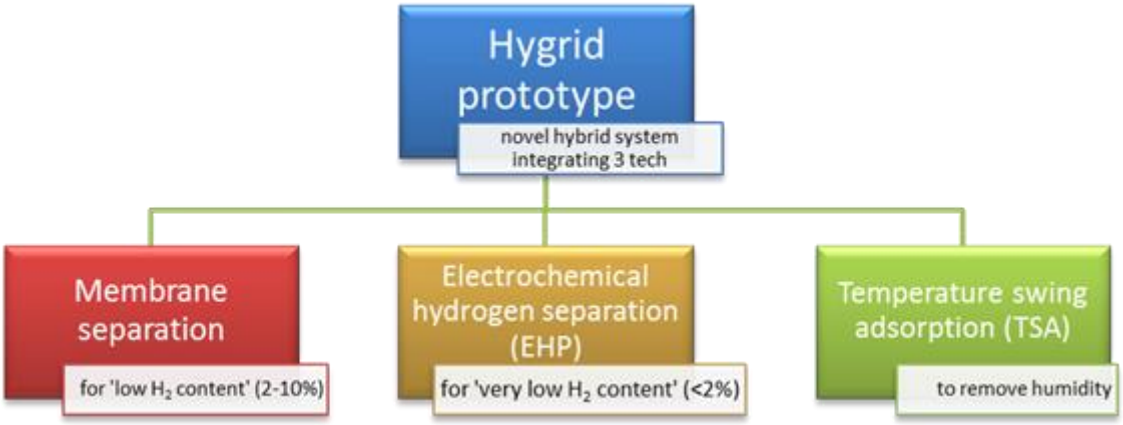


Figure 1. HyGrid concept

The new separation & purification system will increase the value of hydrogen blended into the natural gas grid, improving the economics of central hydrogen production from excess renewable energy couples with natural gas grid injection. In addition, it will reduce cost, and therefore increase the use of hydrogen from very dilute hydrogen streams in energy and transport applications. On the other side, further applications could be found in separating hydrogen from mixtures produced in chemical or biological processes, where it otherwise would be used to generate heat or even be vented.

Project objectives

The HyGrid project will develop, build and demonstrate at industrial relevant condition a novel advanced high performance, cost effective separation technology for the direct separation of pure hydrogen from natural gas grids. In particular, by combining the three different technologies (membrane separation, electrochemical separation and temperature swing adsorption) the total cost of hydrogen recovery will be decreased. The project targets a pure hydrogen separation system with power and cost of < 5 kWh/kg_{H2} and < 1.5 €/kg_{H2}. The pilot will be designed for the separation and purification of >25 kg/day of hydrogen (ISO 14687).

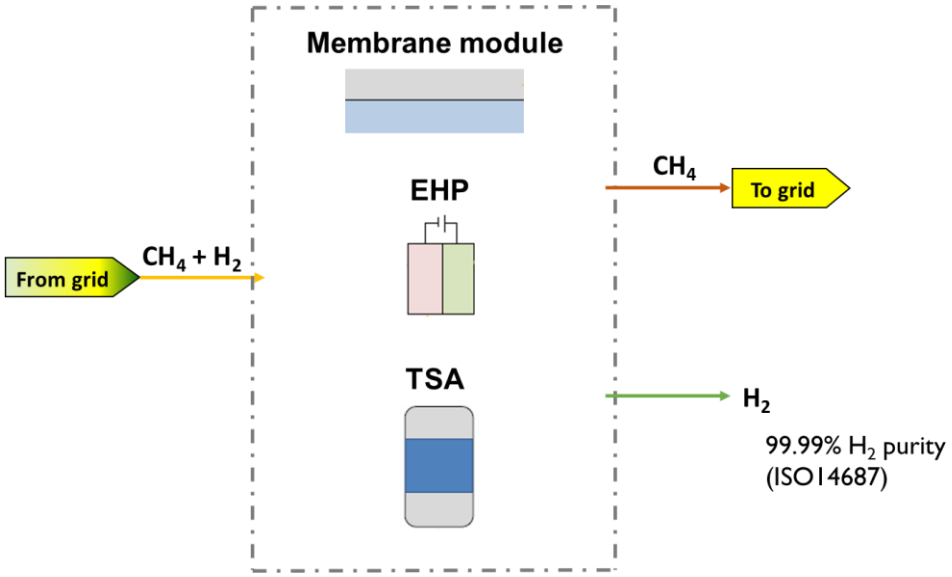


Figure 2. System schematic layout

The main objectives of the HyGrid project are:

- Design, develop, demonstrate and optimise an advanced hydrogen separation system for the production of at least 25 kg/day of hydrogen as per ISO 14687 from low (2-10%) and very low (<2%) H_2 blends in natural gas grids
- Development of stable, high performance and long durability membranes for hydrogen recovery from low (2-10%) hydrogen content streams.
- Development of more stable sealing methods for the membranes at moderate temperatures and reductive atmospheres.
- The further development of EHP for hydrogen recovery from very low (<2%) concentration streams.
- The further development of TSA for water removal from hydrogen/water streams.

- The integration of the new membranes, TSA and EHP in novel hybrid system to achieve high recoveries with low energy penalties.
- Energy analysis of the new HyGrid technology on different scenarios:
  - recovery of H_2 from low concentration streams (2% -10%) up to 99.99% H_2 purity (ISO14687) in the whole range of pressures of the NG grid.
  - Different configurations/combinations of the three separation technologies
- The validation of the novel hybrid system at prototype scale (TLR 5)
- The environmental analysis through a Life Cycle Assessment of the complete chain.
- Dissemination and exploitation of the results.

Partnership

The HyGrid consortium consists of 7 European organizations from 4 countries (Netherlands, Spain, Italy and Switzerland). HyGrid gathers the complete chain of expertise reaching the critical mass necessary to achieve the objectives of the project. The consortium brings together multidisciplinary expertise of material development (electrochemical separation, sorption and membranes), chemical and process engineering, modelling (from thermodynamics to unit operation modelling to system integration), membranes modules and reactors development, LCA and industrial study, innovation management and exploitation.

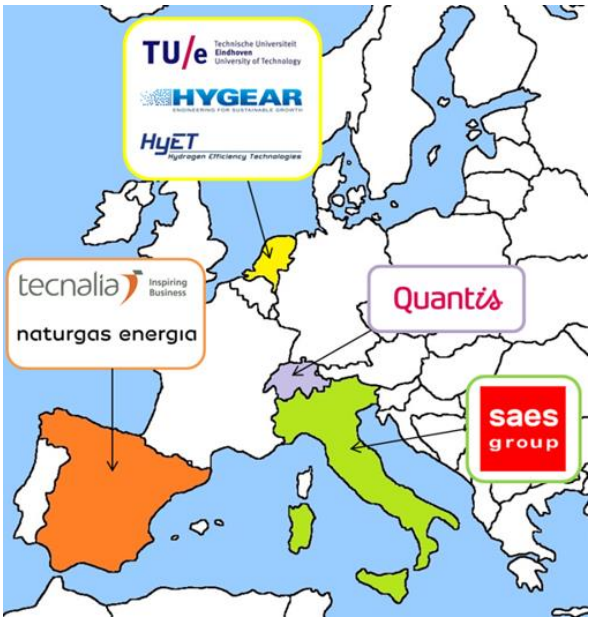


Figure 3. European partnership in HyGrid

Project structure

The HyGrid project structure is subdivided in ten work packages (see the simplified scheme below) following the focus on the development of novel (longer and more stable) membranes for H_2 separation, electrochemical separation and TSA for hydrogen separation from natural gas grids. Furthermore, the project will give a robust proof of concept, validation and assessment of the novel hybrid separation technology. The synergies

between the partners are also visible in the scheme. Therefore, the work structure is based on the following work packages

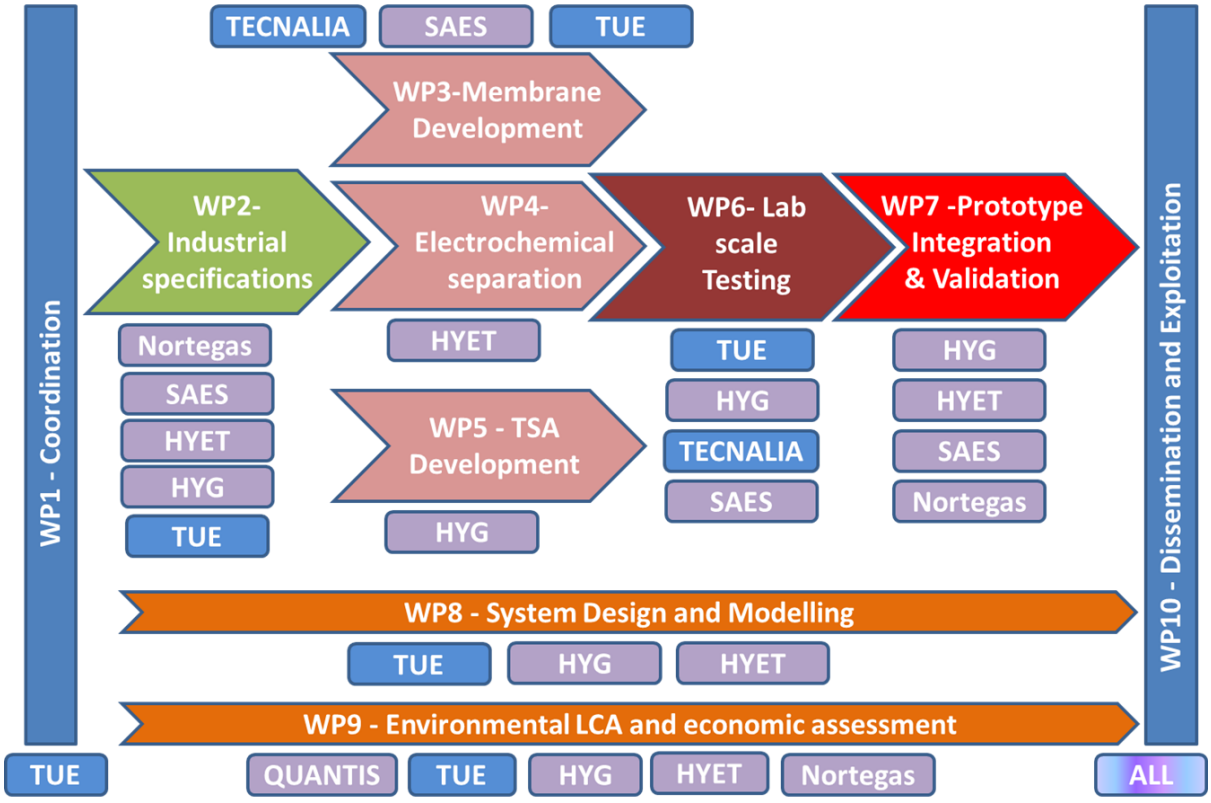


Figure 4. Work structure and synergies between partners.

Latest news from the project

The latest news on different WP activities are now reported:

Membranes development

In HyGrid project, TECNALIA is developing supported Palladium - silver (Pd-Ag) and composite alumina carbon molecular sieves membranes (Al-CMSM). Pd based membranes have very high permeation and selectivity towards hydrogen and their best temperatures of permeation are in the range of 300 to 500 °C. On the other hand, Al-CMSM membranes have lower permeance and selectivity, but they can permeate hydrogen at low temperatures (below 100 °C) where Pd membranes can be damaged; in addition, the Al-CMSM can be prepared from cheap polymeric precursors.

Thin Pd-Ag supported on porous ceramic supports show higher hydrogen permeation than Pd-Ag supported on porous metallic supports. However, they are less mechanically strong than the metallic ones and they are more difficult to integrate in a metallic module

because stable and leak-free connections between ceramic supports and metallic tubes are difficult to obtain. Long term test has shown that leaks/defects appear mainly in these connections with higher probability than in other parts of the membrane, especially when the porous ceramic supports have thin walls and it is not possible to apply a high enough torque in the connection to obtain leak-free joints. To avoid this problem, HyGrid will use Pd-Ag 50 cm long membranes (5 µm thick) supported onto tubular finger like alumina supports (14/7 mm o.d/i.d.) to increase the torque capacity of the support and decrease the number of joints (Figure 5).

Figure 5. Finger type Pd-Ag membrane supported on porous alumina support.

On the other hand, Al-CMSM membranes, despite not having as high hydrogen permeation as the Pd based membranes (one to three orders of magnitude lower, respectively at more than 300 °C), they are cheaper and can operate at low temperature (< 300 °C), including ambient temperature, where the sealing is not a main issue. Recently, composite Al-CMSM of 40 cm long have been prepared and will be studied for the separation of H_2 in NG at high pressures (Figure 6).

Figure 6. Al-CMSM 40 cm long.

Electrochemical hydrogen separation development

The main objective of this task is the development of a hydrogen purifier (EHP) prototype for the recovery of the hydrogen from low concentration streams (H_2 ≤2 %) to be integrated in the final hybrid separation/purification prototype.

In the previous months, progress has been made in tests on single cell EHP stacks. The water management in the cell is improved to obtain stable performance of the cell over a time span of approximately one or two days.

However, in the previous newsletter, we mentioned that the cell-to-cell performance uniformity needed to be improved. Many different tests have been performed on 5-cell EHP stacks in the meantime, to finetune the optimal conditions for temperature, pressure and relative humidity of the EHP. These tests have shown that the performance of multicell stacks can be better than single cell stacks. However, cell-to-cell performance needs to be homogeneous, which is challenging with the low concentration of hydrogen in the feed gas.

In the previous period, it was concluded that the first prototype of the EHP needed improvement, especially on the pressure drop of the gas flow through the EHP. The engineering of the new prototype is now being finalized. The modelling of the EHP has already finished a few months ago. The results of the modelling work have been used in the engineering of the second prototype.

In the upcoming months, the second prototype will be tested and integrated in a module that can be incorporated in the overall HyGrid system. HyET will continue to cooperate with HyGear to successfully deliver the EHP module for the system this year.

Temperature Swing Adsorption development

HyGear has finished the assembly of the TSA system at its premises. Debugging of the system software has started. Test results are expected in the next period.

Lab scale testing

A Pd-Ag ceramic supported membrane, depicted in Figure 7, with a 10/7 mm support was tested in presence of sweep gas comparing nitrogen and steam as sweep gas. At the beginning the tests were performed at 400 °C changing the total retentate pressure from 5 to 2 bar while keeping the total permeate pressure at 1 bar. The feeding mixture was 90% CH_4 and 10% H_2. As previously observed, increasing the sweep gas flow rate leads in a lower hydrogen permeation over an upper limit due to a raise in the pressure drop in the porous support. Furthermore, the in-house developed model to evaluate the permeance have also predicted the expected permeance while taking into account the concentration polarization in the retentate and permeate side and the mass transfer limitation in the porous support.

The maximum hydrogen volume flow rate at 4 bar pressure difference and with the highest sweep gas flow rate, is 250 ml/min while for the same conditions in presence of steam is 150 ml/min.

In addition, TGA tests on alumina supports were carried out for checking any effect of steam at different temperatures. The setup is depicted in Figure 8.

Figure 7. Pd-Ag membrane with an effective thickness layer between 2-3 µm, outer diameter of 10 mm and inner diameter of 7 mm.

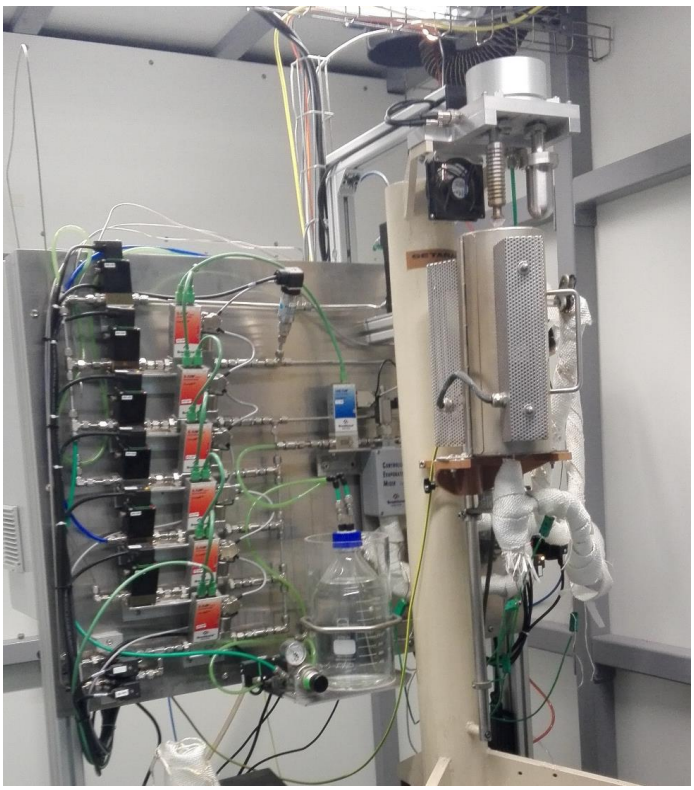


Figure 8. Thermal gravimetric analysis setup.

The other lab scale task was testing the membranes at high pressure; hence, high pressure experiments with carbon membranes were performed for a proper understanding of the hydrogen permeance at high pressure. At the beginning pure gas tests were performed and later on, mixtures with 10% H_2 – 90% CH_4.

System modelling and simulation

A proper membrane model which includes concentration polarization in the retentate, permeate and mass transfer limitation in the porous support was proposed and validated in comparison with experimental results at different operating conditions. Several aspen simulations were carried out for a proper optimization of the hybrid configuration for the prototype selection.

Figure 9. Hybrid system composed of membrane module, electrochemical hydrogen compressor and temperature swing adsorption.

Prototype development

The design of the HyGrid prototype has been continued. Discussions with the different partners to integrate the TSA module with the membrane module and the EHP module were continued. Based on the discussions HyGear prepared the concept process flow diagram of the integrated HyGrid prototype for testing at HyGear. Currently, we are building the stream tables to determine the impact on auxiliaries costs of several process flow configurations and we started with the design of the balance of plant for the different modules. It is expected to finish the selection of balance of plant in the next period. Early 2019 it is expected to start with the assembly of the prototype at HyGear.

Environmental and economic assessment

The environmental and economic assessment of the new hydrogen recovery systems developed within the HyGrid project will also be evaluated. The aim is not only to compare the developed technologies to current hydrogen recovery systems, but also to guide the design of the investigated technologies towards more environmentally friendly solutions. The core methodology that will be used to achieve this is life cycle assessment (LCA), a quantitative environmental assessment tool which estimates the environmental impacts of products or services looking at their entire life cycle as shown in Figure 10 below.

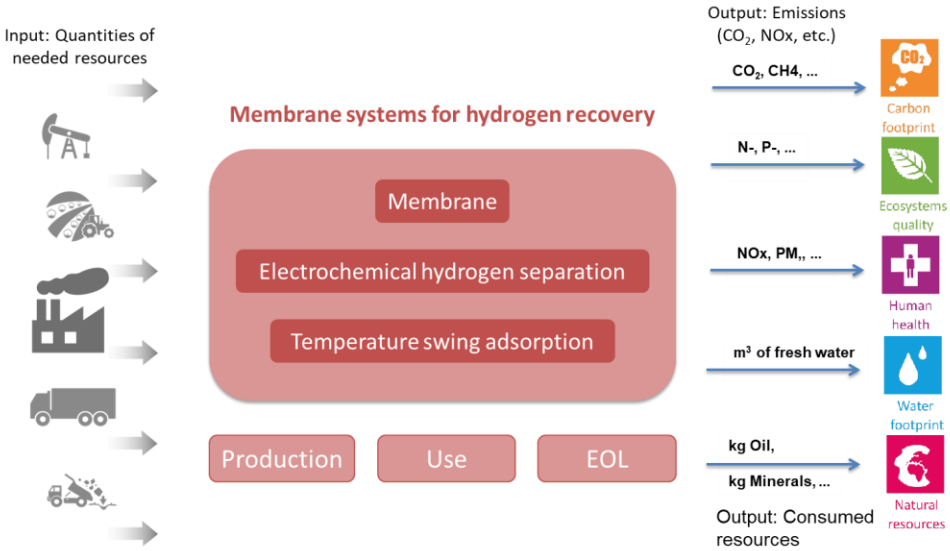


Figure 10. LCA approach.

In the first 12 months, the first task of this assessment was completed which involved developing the framework for the environmental and economic assessment. In technical LCA terms, this corresponds to developing the goal and scope of the study which involves clearly defining what different systems will be analysed, what system boundaries and functional unit will be used for the study and what reference technology the HyGrid system will be compared to.

The functional unit for this study was determined to be: "the recovery of 1 kg of hydrogen with a purity of at least 99.97% from an average European natural gas grid". Pressure swing adsorption (PSA), which had been identified as a suitable reference technology in the first period of the project, was further scrutinized. The results of this further analysis showed that PSA is not suitable for separating hydrogen from methane. Therefore, there are no real comparable systems to the HyGrid system.

During the last 6 months, data collection for the HyGrid system as well as its modelling was performed. The preliminary LCA results were presented at the M24 meeting. They show a high contribution of the electricity use of the system in all environmental impact indicators. The most important components in terms of impacts are so far the TSA and the EHP, due to the high metal consumption of these two components. In the next period of this project, the modelling will be refined to have a more detailed representation of the components as well as to include their end-of-life.

Highlights

HyGrid Workshop 2018
Exploitation Workshop on Flexible Hybrid Separation System for H_2 Recovery from Natural-Gas Grids
May 17th, 2018 in Lainate, Italy

SAES Getters SpA hosted the HYGRID Exploitable Workshop on flexible hybrid separation for H_2 recovery from NG grids. The focus was on hydrogen generation, distribution and transportation, challenges of distributing hydrogen into the NG networks and the purity requirements. The workshop was part of the first transfer of knowledge event of the HyGrid project, contributing to the increase of knowledge, and competitiveness of the hydrogen economy in the EU community. Presentation are available in the HyGrid website (https://www.hygrid-h2.eu/content/publications).

PROMECA Workshop 2018:
Membrane and Membrane Reactors
December 4th, 2018 in Eindhoven, The Netherlands

Eindhoven University of Technology will host the 3rd PROMECA Workshop on Membrane and Membrane Reactors. The objective of this one-day workshop in Eindhoven (The Netherlands) is to present the state of the art of membranes for gas separation, catalysts and membrane reactors, indicating the industrial specifications and the modelling and design of innovative plants based on membrane reactors technologies. It will give and overview of the actual research results on membranes development for gas separation (i.e. O_2, H_2,) based on organic and inorganic membranes, catalyst and catalytic membranes, membrane reactors for H_2 production from different sources as well applications of membrane reactor in m-CHP systems. The workshop also includes a visit in Eindhoven at the laboratories of the Inorganic Membranes and Membrane Reactors group. Please visit PROMECA website for updates (www.promecaproject.com).

The workshop will be part of the third transfer of knowledge event of the PROMECA project, contributing to the increase of knowledge, skills, and competitiveness in the European Community of membrane reactors.

14th International Conference on Catalysis in Membrane Reactors July 8 - 11, 2019 in Eindhoven, The Netherlands

On July 8 – 11, 2019, the 14th International Conference on Catalysis in Membrane Reactors (ICCMR14) will be held in Eindhoven, The Netherlands. The ICCMR14 will be organized by the Eindhoven University of Technology.

The aim of the ICCMR conferences is to promote the research and progress in the area of catalytic membrane systems by bringing together academic scientists and industry working in the membrane, catalysis and process engineering fields. The meeting will highlight recent developments, bring new ideas, help making contacts and create a platform for discussion between academics and practitioners. We strongly believe that all the participants will have a great opportunity to make beneficial contacts and exchange ideas. The conference is addressed also to young researchers who will have a chance to interact closely with senior scientists. Please visit the conference website for updates (https://www.iccmr14.com/)).

Dissemination activities, publications and presentations:

HyGrid public presentations as well as open access articles and public reports are available online in the dissemination section of the project website: www.hygrid-h2.eu.

Peer reviewed articles:

1. Margot A. Llosa Tanco, David A. Pacheco Tanaka. Recent Advances on Carbon Molecular Sieve Membranes (CMSMs) and Reactors. Processes 2016, 4, 29; doi:10.3390/pr4030029.

2. A.M. Gutierrez, J.R. Arraibi, M.A. Llosa Tanco, J. Zúñiga, J.L. Viviente, L. García Gómez. Development of carbon molecular sieve Membranes for the use of renewable gases, biomethane and hydrogen in natural gas networks. Proceeding of the International Gas Union Research Conference 2017 (IGRC2017). Rio de Janeiro, Brazil (24-26/05/2017).

Other dissemination activities:

1. M. Nordio, F. Gallucci, M. van Sint Annaland, V. Spallina. *Flexible Hybrid separation system for Hydrogen recovery from Natural gas Grids*. Dutch membrane meeting (2016). Poster

2. Naturgas. *Una industria energéticamente sostenible.* Newspaper El Correo – Innovation section. Bilbao, Spain (1st June 2016).

3. Martijn J.J. Mulder, Peter J. Bouwman. The need for High Temperature Proton Exchange Membranes for electrochemical hydrogen purification and compression. EMEA workshop 2016. Bad Zwischenahn, Germany (27-29/06/2016). Poster.

4. A.M. Gutierrez, Flexible Hybrid separation system for H2 recovery from Natural Gas Grids (HyGrid). GERG Meeting with DG ENERGY, Brussels, Belgium (06/02/2017). Oral

5. F. Gallucci, J.L. Viviente. Flexible Hybrid separation system for H2 recovery from NG Grids. Third European Workshop on Membrane reactors: Membrane Reactors for Process Intensification (MR4PI2017). Villafranca di Verona, Italy (9-10/03/2017). Poster.

6. Marco Succi, Giorgio Macchi. Pd Supported Membrane Hydrogen Purifier: a comparison with other technologies. Third European Workshop on Membrane reactors: Membrane Reactors for Process Intensification (MR4PI2017). Villafranca di Verona, Italy (9-10/03/2017). Poster.

7. A.M. Gutierrez. Hidrógeno en redes de gas natural. Fronteras Tecnológicas en Generación de Electricidad, Energías Renovables e Hidrógeno Whorshop. Madrid, Spain (26/04/2017). Oral

8. A.M. Gutierrez, J.R. Arraibi, M.A. Llosa Tanco, J. Zúñiga, J.L. Viviente, L. García Gómez. Development of carbon molecular sieve Membranes for the use of renewable gases, biomethane and hydrogen in natural gas networks. International Gas Union Research Conference 2017 (IGRC2017). Rio de Janeiro, Brazil (24-26/05/2017). Poster.

9. M. Succi, G. Macchi, E. Fernandez, J. Melendez, J. L. Viviente, D.A Pacheco Tanaka. Advancement in Palladium Membranes Hydrogen Purification. 6th European PEFC and Electrolyser Forum. Lucerne, Switzerland (4-7/07/2017). Poster

10. D.A. Pacheco Tanaka, M.A. Llosa Tanco, J. Medrano, J. Melendez, E. Fernández, M. Nordio, F. Gallucci. Preparation and hydrogen permeation studies of ultra-thin Palladium (≈1 micrometer) and carbon membranes from mixtures containing low concentration of hydrogen (< 30%). 13th International Conference on Catalysis in Membrane Reactors (ICCMR13). Houston (Texas), USA (10-13/07/2017). Oral presentation: Key note.

11. M. Nordio, M. Van Sint Annaland, F. Gallucci, V. Spallina, M. Mulder, L. Raymakers, P. Bouwman. Electrochemical Compressor for Hydrogen Separation in a Small-Scale Hybrid System. 13th International Conference on Catalysis in Membrane Reactors (ICCMR13). Houston (Texas), USA (10-13/07/2017). Oral presentation.

12. M. Nordio, J. Meléndez, E. Fernández, M. Van Sint Annaland, D.A. Pacheco Tanaka, F. Gallucci. Ultra-thin palladium-silver membranes for pure hydrogen production and separation: modelling and effect of sweep gas. 13th International Conference on Catalysis in Membrane Reactors (ICCMR13). Houston (Texas), USA (10-13/07/2017). Oral presentation.

13. M. Nordio, J. Meléndez, D.A. Pacheco Tanaka, M. Mulder, P. Bouwman, L. Raymakers, M. Van Sint Annaland, F. Gallucci. Hybrid separation system for hydrogen recovery from natural gas grids. 10th World Congress of Chemical Engineering (WCCE10). Barcelona, Spain (1-5/11/2017). Oral presentation.

14. A. Bos, M. Mulder, P. Veltman. Electrochemical Hydrogen Compression as New Disruptive Technology in Hydrogen Purification and Storage. European Hydrogen Energy Conference 2018 (EHEC). Malaga, Spain (March 14th, 2018). Oral presentation.

15. F. Gallucci. HyGrid Project: Objectives and scope. HyGrid Workshop 2018: Exploitation Workshop on Flexible Hybrid Separation System for H2 Recovery from Natural-Gas Grids. Lainate (Mi), Italy (May 17th, 2018). Oral presentation.

16. A. Gutierrez. The challenge of distributing Hydrogen into the natural gas network. HyGrid Workshop 2018: Exploitation Workshop on Flexible Hybrid Separation System for H2 Recovery from Natural-Gas Grids. Lainate (Mi), Italy (May 17th, 2018). Oral presentation.

17. A. Pacheco, G. Macchi. Hydrogen separation using membranes. HyGrid Workshop 2018: Exploitation Workshop on Flexible Hybrid Separation System for H2 Recovery from Natural-Gas Grids. Lainate (Mi), Italy (May 17th, 2018). Oral presentation.

18. L. Raymeakers. Electrochemical hydrogen compression and purification. HyGrid Workshop 2018: Exploitation Workshop on Flexible Hybrid Separation System for H2 Recovery from Natural-Gas Grids. Lainate (Mi), Italy (May 17th, 2018). Oral presentation.

19. L. Roses. Design of the HyGrid prototype. HyGrid Workshop 2018: Exploitation Workshop on Flexible Hybrid Separation System for H2 Recovery from Natural-Gas Grids. Lainate (Mi), Italy (May 17th, 2018). Oral presentation.

20. R. Swanborn. Electrochemical hydrogen compression coming to age. Hydrogen + Fuel Cells North America 2018. Anaheim (CA), USA (September 26th, 2018). Oral presentation.

Upcoming events

November 2018	FCH JU Programme Review Days 2018 Brussels, Belgium http://www.fch.europa.eu/
December 4th, 2018	PROMECA workshop 2018: Membranes and Membrane Reactors, Eindhoven (The Netherlands). http://promecaproject.com/
April 24 – 26, 2019	14th HYdrogen - POwer THeoretical and Engineering Solutions International Symposium (HYPOTHESIS XIV), Foz do Iguaçu (Brazil) http://www.hypothesis.ws/
June 2 – 7, 2019	8th World Hydrogen Technology Convention (WHTC 2019) Tokyo, Japan http://whtc2019.jp/
June 16-20, 2019	8th International Zeolite Membrane Meeting (IZMM2019), Lulea (Sweden). https://www.ltu.se/research/subjects/Kemisk-teknologi/Konferenser/IZMM2019?l=en
July 8-11, 2019	14th International Conference on Catalysis in Membrane Reactors, Eindhoven (The Netherlands). https://www.iccmr14.com/
June 22 – 25, 2020	23rd World Hydrogen Energy Conference (WHEC 2020), Copenhagen (Denmark)

HyGrid in figures:

- *7 partners (2RES, 2 IND, 3 SME)*
- *4 countries*
- *2,847,710 € project (2,527,710 € EU funded)*
- *Start May 2016*
- *Duration: 36 months*

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More information on HyGrid (including a non-confidential presentation of the project) is available at the project website: www.hygrid-h2.eu

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Disclosure:

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