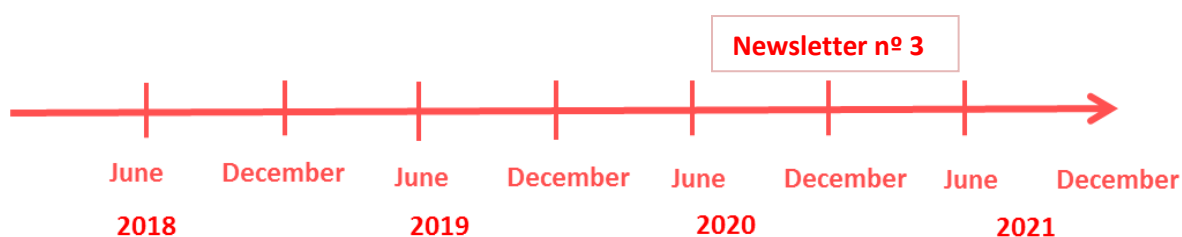


Advanced MEMBranes and membrane assisted procEsses for pre- and post- combustion CO₂ captuRe

Newsletter – Issue 3 –2020



Editorial

Welcome to this third MEMBER newsletter. MEMBER is a four-year project aiming to demonstrate state-of-the-art CO₂ capture technologies in an industrially relevant environment. MEMBER will scale-up and manufacture advanced materials and prove their added value in terms of sustainability and performance. It targets three advanced solutions based on: Innovative MOF -MMMs for pre- and post- combustion CO₂ capture in power plants, and an intensified reforming process combining high temperature solid CO₂ sorbent and dense Pd membranes for pure H₂ production with integrated CO₂ capture (MA-SER).

The present newsletter presents the progress of the project and highlights information related to the R&D fields addressed in the project. We hope you will find the info in this newsletter interesting. On our website <https://member-co2.com/> you will find public presentations, all the public deliverables of the project and more interesting news. Stay tuned!

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What is MEMBER?

The concept

Currently, more than 80% of global primary energy use is fossil based. Over the last decade, 85% of the increase in global use of energy was fossil based. In the transition to a fully low-carbon economy, the Carbon Capture and Storage (CCS) technology is one of the key ways to reconcile the rising demand for fossil fuels, with the need to reduce greenhouse gas emissions. Globally, CCS is likely to be a necessity in order to keep the average global temperature rise below 2 °C.

The main drawback of including CCS in power generation or other industrial sectors is related to the energy consumed by the separation processes needed to achieve low carbon emissions (e.g. heat for solvent regeneration). This energy requirement results in a higher fuel consumption and higher fuel cost, leading to reduced overall net efficiencies. Together with the increased CAPEX, due to additional equipment (separation processes or chemical reactors), it determines the substantial increase of the cost of electricity or of an industrial product when CO₂ capture is included.

In order to reduce this energy penalty, MEMBER develops three advanced CO₂-capture solutions using less energy. These technologies are:

- Innovative Mixed-Matrix-Membranes (MMM) filled with Metal-Organic-Frameworks (MOF) for pre- and post- combustion CO₂ capture in power plants;
- An intensified reforming process combining catalytic reforming with a high temperature solid CO₂-sorbent and dense Pd-membranes for pure H₂ production with integrated CO₂-capture (Membrane Assisted Sorption Enhanced Reforming, MA-SER).

MEMBER aims to demonstrate these state-of-the-art capture technologies in an industrially relevant environment. To achieve this, MEMBER will scale-up and manufacture advanced materials and will prove their added value in terms of sustainability and performance under industrially relevant conditions (TRL 6) in novel membrane based technologies. These new technologies will outperform current technologies for pre- and post-combustion CO₂ capture in power plants as well as H₂ generation with integrated CO₂ capture and meet the targets of the European SET plan. In both cases, a significant decrease of the total cost of CO₂ capture will be achieved. MEMBER targets CO₂ capture technologies that separate >90% CO₂ at a cost below 40€/ton for post combustion and below 30€/ton for pre-combustion and H₂ production.

The developments within MEMBER have started using the best materials and technologies developed in three former FP7 projects, ASCENT, M⁴CO₂ and FluidCELL. In particular, special attention will be paid to the scale-up and improvements of manufacturing



processes of key materials and products such as Metal-Organic-Frameworks (MOFs), polymers, membranes and sorbents.

Table 1. Performance targets for the MEMBER prototypes

	Technology	CO ₂ Capture [%]	Capture cost [€/ton]	Demo site
Pre-combustion (Power plant)	MMM	> 90	< 30	CENER
Post-combustion (Power plant)	MMM	> 90	< 40	GALP
H ₂ with integrated CO ₂ capture	MA-SER	> 90	< 30	IFE-HYNOR

Project objectives

The key objective of the MEMBER project is the scale-up and manufacturing of advanced materials and their demonstration at industrially relevant conditions (TRL6) in novel membrane-based technologies that outperform current technologies for pre- and post-combustion CO₂ capture in power plants as well as H₂ generation with integrated CO₂ capture and meet the targets of the European SET plan.

Three different technological solutions involving advanced materials will be developed and demonstrated at three different end user's facilities:

- Advanced Mixed Matrix Membranes (MMMs) for pre- and post-combustion CO₂ capture in power plants (H₂/CO₂ & CO₂/N₂ respectively)
- A combination of metallic hydrogen membranes and CO₂ sorbent integrated into an advanced Membrane Assisted Sorption Enhanced Reforming (MA-SER) process for pure H₂ production with CO₂ capture.

The main objectives of the MEMBER project are:

- Increasing the manufacturing readiness level (from MRL 4-5 to MRL 6) of a portfolio of materials for the production of Mixed Matrix Membranes for pre- and post-combustion CO₂ capture in power plants (H₂/CO₂ and N₂/CO₂ separation).
- Increasing the manufacturing readiness level (from MRL 4-5 to MRL 6) of hydrogen membranes, reforming catalysts and CO₂ sorbents materials, and integrating them into an advanced Membrane Assisted Sorption Enhanced Reforming (MA-SER) process for pure H₂ production with CO₂ capture.
- Development of a software tool to simulate MEMBER components, the processes and CO₂ capture energy performance.
- Design and construction of 3 prototypes for CO₂ capture for testing of the developed materials in relevant operating conditions at TRL6.

- Demonstration of the MEMBER systems and related business models in 3 representative demonstration sites across Europe, covering different sectors, membrane-based technologies and CO₂ containing streams:
 - Prototype A targeted for pre-combustion in a gasification power plant using MMM at the facilities of CENER (BIO-CCS).
 - Prototype B targeted for post-combustion in power plants using MMM at the facilities of GALP.
 - Prototype C targeted for pure hydrogen production with integrated CO₂ capture using (MA-SER) at the facilities of IFE-HyNor.
- Quantification of the environmental impacts of the proposed holistic solutions through life cycle assessment.
- Exploitation of the results including the definition of a targeted and quantified development roadmap to bring the technologies to the market.
- Overcoming the CCS market barriers with an ambitious set of CCS solutions.

Partnership

The consortium brings together multidisciplinary expertise on the entire value chain: material development (MOFs, polymers, sorbents and catalysts), membrane development (MMMs, Pd based membranes); chemical and process engineering, modelling (from thermodynamics to unit operation modelling to system integration), membrane modules and reactors development, recycling, LCA and industrial study, innovation management and exploitation. It is composed of 17 partners from 9 countries: 6 RTO/HES and 11 SMEs/INDs (65%) it is an industrial oriented consortium, including 7 innovative SMEs (41%) and 4 Large industries (24%).

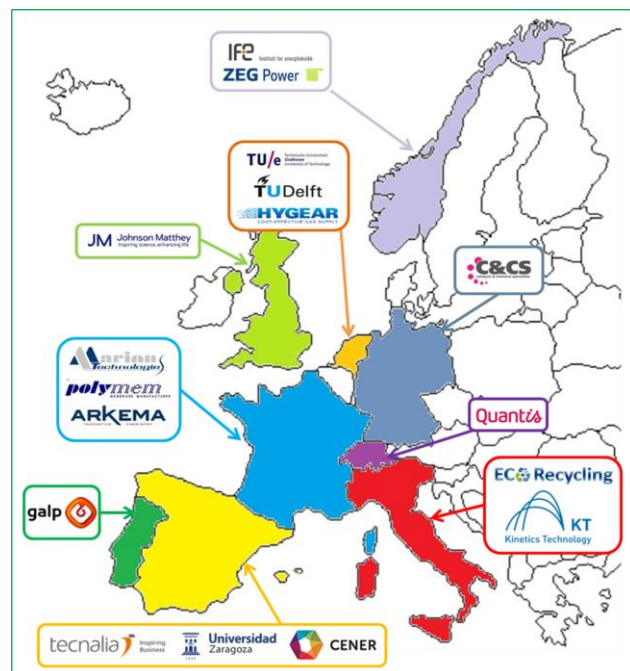


Figure 1. European partnership in MEMBER

Project structure

The MEMBER project structure is subdivided in ten work packages following the focus on the development of the CO₂ capture processes. Furthermore, the project will demonstrate the capture technologies in industrially relevant environment. Therefore, the work structure is based on the following work packages.

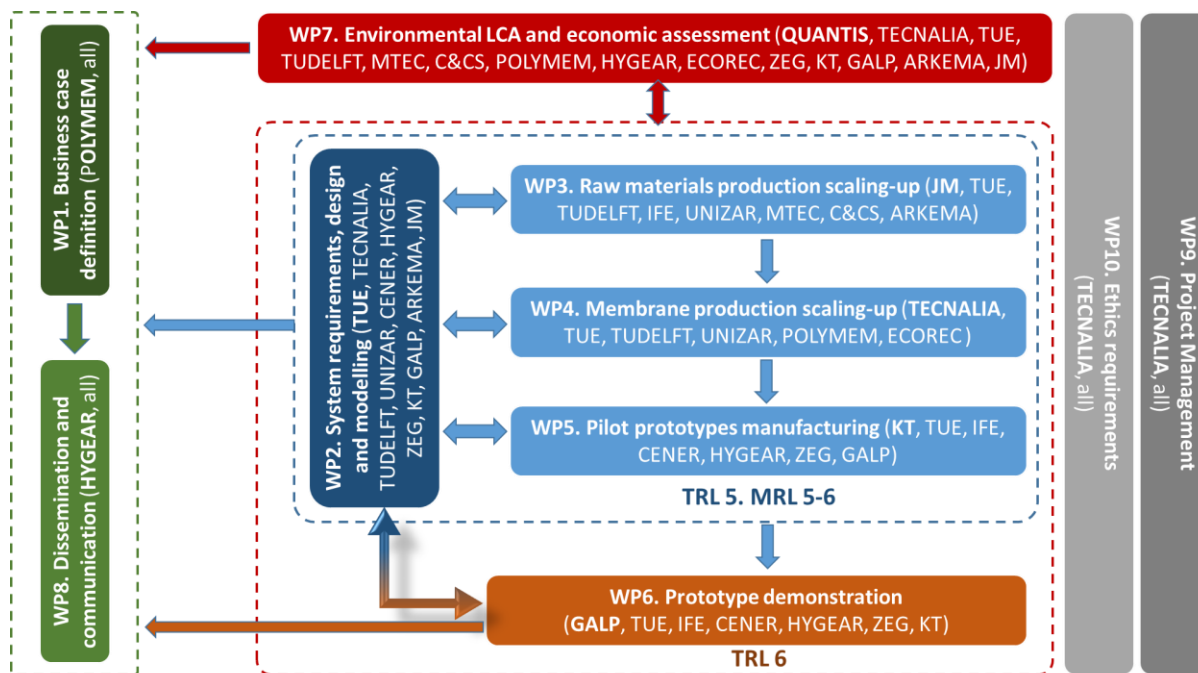


Figure 2. Work structure and synergies between partners.

Latest news from the project

The latest new on different WP activities are now reported:

Business case definition

WP1 is setting the foundation for effective development and exploitation of MEMBER project results into the market. Main objectives of WP1 are to characterise the market, to assess the outstanding features of exploitable results, to manage and protect our intellectual properties; to identify appropriate business models for every MEMBER developed technologies and to set a plan for commercialisation and market uptake; finally, to characterise the different development risk and identify solutions to mitigate them.

From the beginning of the project the main following actions have been done:

- market analysis;

- initial risks have been listed and solution to mitigate them proposed.
- Value proposition of MEMBER technologies, including a preliminary identification of all the key exploitable results have been done.
- Several business models related to different MEMBER technologies have been defined.
- Go to market strategy has just started for all the previous defined business cases.

During the first period, WP1 partners did a classical top-down analysis of the market using global data but also, we did a bottom-up analysis from the information of the MEMBER partners collected through a survey. From the top-down analysis, we confirmed the Carbon capture market is enormous. The size of Carbon Capture market is 4 Giga ton of CO₂ per year to reach Paris Agreement and to limit to 2°C the global warming. Using numbers coming from the M4CO₂ project saying 1 m² of membrane can concentrate and capture 2 kg/h of CO₂ i.e. 17 ton_{CO₂}/y, the 4 Gton requests 235,000,000 m².

On the other side, from the bottom-up analysis, we defined the 3 main products developed during the Member project:

1. CO₂ capture and H₂ production units in pre-combustion based on H₂/CO₂ separation by Mixed Matrix Membrane
2. CO₂ capture units in post-combustion based on CO₂/N₂ separation by Mixed Matrix Membrane
3. CO₂ capture and H₂ production units based on MA-SER process assisted by Pd membrane

In addition, we found potential customers and define initial business cases corresponding to small emitters which are not subjected to CO₂ allowances but could be interested by MEMBER products in order to save costs on the supplying of liquified CO₂. CO₂ using MEMBER technology should be at 40€/ton when liquified CO₂ cost is between 100 and 150€/ton.

During the second period, large CO₂ emitters, which were not considered initially because the maturity of our MEMBER technologies was too low and far from these large-scale perspectives, have been re-considered in order to cover all the markets sizes for longer term “go to market strategy”. Business cases matrix was improved with several larger cases.

An in-depth analysis of several identified business cases lays the foundation for defining the four business models covering exploitation schemes for all three MEMBER processes for specific industry sectors or niche applications. Further evaluation of technical and economic aspects is ongoing to decide over viability of these business cases. Based on this work, 4

main business models for a variety of possible constellations of CO₂ production and capturing have been defined.

Finally, a template for “Preliminary commercialization & market strategy” has been proposed to the partners in order to document the commercialization routes and strategy to market. This template has been discussed and approved by partners and is going to be filled by the beneficiaries of business models in the next and final period of the project.

The main conclusions from the market analysis are:

1. The CCS market is in its infancy.
2. Carbon prices need to increase considerably to make a sustainable business case.
3. Launching customers will be small-size CO₂-emitters, up to about 25000 T/y having a need for CO₂ in their processes. For instance, sodium carbonate producers, water treatment (pH regulation), and food industries.

System requirement, design and modelling

Within this WP the system requirements have been defined and the different membrane modelling as well as the reactor and MMMs system modelling have been completed. Regarding the membrane modelling main achievement are detailed here after.

The steps for the purification of hydrogen using metallic supported Pd-Ag membranes in a membrane module has been completed. After the identification of the resistances present in the purification process, the significance of the individual resistances was determined to simplify the mathematical model for the description of the membrane module permeation. Three distinct mass transfer resistance/ steps were identified to contribute to overall membrane flux; *(i)* External mass transfer at retentate side, bulk-to-membrane surface diffusion, *(ii)* Dense membrane lattice diffusion, *(iii)* Mass transfer resistance due to porous media, molecular and/or Knudsen diffusion. Finally, a parametric study was performed to determine the membrane configuration and membrane area required for the target performance parameter set in the CA for the recovery of 10 Nm³/h of hydrogen in the prototype C design.

The modelling of polymeric membranes within the framework of the MEMBER project has been also addressed. The permeation behaviour of polymeric membranes was analysed through the mathematical models considering membrane type, material, geometry, morphology, and configuration. The permeation of flat sheet membrane such as PDMS, Pebax 1657, Polysulfone (Psf) was modelled in pre- and post-combustion processes via mathematical model based on mass balances and transport equations. Further, a sensitivity analysis was performed to investigate the influence of feed concentration on the separation performance of the polymeric membranes. Regarding the gas permeation in HF membranes,



the preliminary studies were carried out for the membranes composed of Psf as support and Pebax 1657 (3 wt%) as a selective layer.

In addition, studies on the prediction of permeation through mixed matrix membranes comprising ZIF-94 and NH₂-MIL-53(Al) as a filler and Pebax 1657 and 6FDA-DAM polymers as the continuous phase have been performed. Firstly, the permeation through the filler in MMMs was obtained via computational models and then obtained values were applied to predict the permeation of MMMs by ideal and modified analytical models (3-4% deviation from experimental permeability values). Secondly, the thickness and permeability of the interphase layer were estimated by using the modified analytical models by considering the non-idealities in the membrane matrix. The influence of filler loading on MMM permeability was investigated using analytical and computational models. Further, the permeation performance of supported thin mixed matrix membrane were calculated by assuming Knudsen permeation in the porous support and using the acquired permeation data from modified analytical models for the selective layer. Moreover, the permeation performance of MMMs upon different feed conditions (CO₂ partial pressure and temperature) was estimated by using Arrhenius type relations and the experimental CO₂ adsorption data of the membranes at room temperature. As a result, the enthalpy of sorption, activation energy of diffusion, and activation energy of permeation of the MMMs were obtained. Lastly, modelling of module hydrodynamics in HF membranes and the influence of flow pattern (co- and counter-current operation) in high and low flux modules were investigated (Figure 3).

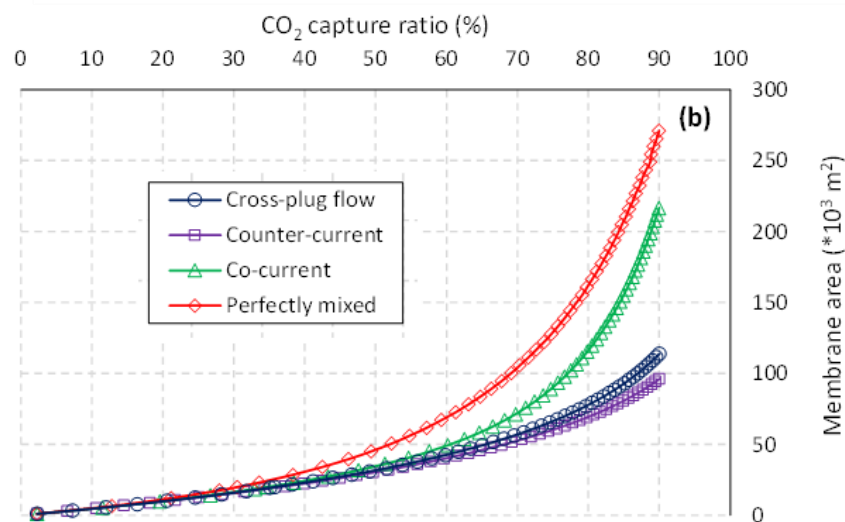


Figure 3. Influence of the flow pattern on the membrane area needed to achieve different CO₂ capture ratio.

The modelling of the reactor for the Membrane Assisted Sorption Enhanced reforming (MA-SER) has been completed. The MA-SER model has been improved for the mathematical description of the MA-SER process with the objective to validate the results from the

Prototype C design and the upscale for large scale hydrogen production. The model allows the user to investigate the operation and design of a (dual) bubbling fluidized bed reactor at isothermal operation conditions (Figure 4). It allows to investigate the effect of a bi-particle mixture of sorbent and catalyst on the performance of the reactor. To have a model that predicts the performance accurately the accurate kinetic expression for both reforming catalyst and CO₂ adsorption must be implemented together with the effect of hydrodynamic of the system. To test the implementation of the kinetics for catalytic and gas-solid reactions the model is validated by transforming the fluidized bed reactor model artificially to a fixed bed reactor, where analytical solutions exist for first order reactions.

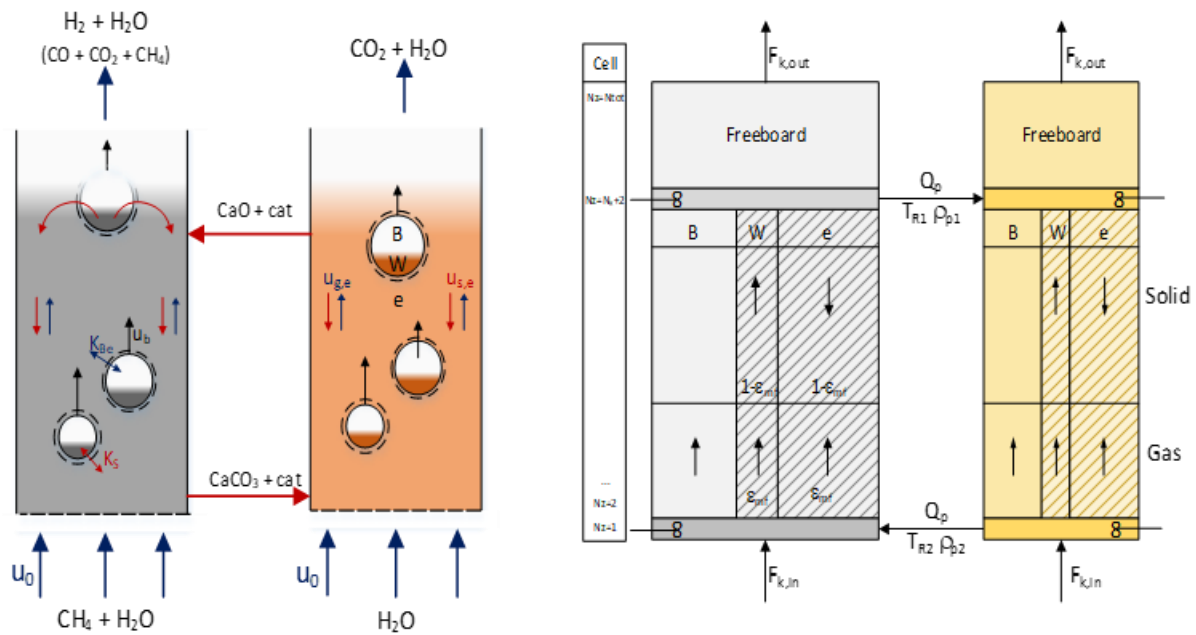


Figure 4. MA-SER reactor system phenomenological model scheme.

Finally, a thorough optimization on the pre- & post-combustion systems based on MMMs has been performed. Firstly, the membrane models for hollow fiber membrane are revised to take the pressure drop into consideration (to optimize module length) and validated by comparing the calculated results with the experimental data in literature and the MEMBER project. Secondly, the post-combustion CO₂ capture process is optimized by the Genetic Algorithm (GA) approach and further complemented by a sensitivity analysis. The GA approach is a new emerging methodology to optimize membrane separation processes. The large number of variables to be optimized in concert may lead to local minima solutions with classical optimization routines, while by the GA approach a global solution is sought, in this project for a minimum energy consumption. Around this minimum the influence of varying the operating conditions on the energy consumption is investigated, while still satisfying the fixed boundary conditions of CO₂ purity and recovery. Further, the pre-combustion CO₂ capture process, with less degrees of freedom (less variables), is analysed and optimized with sensitivity analysis. The trends observed in the sensitivity analysis confirmed the optimal operating conditions.

Core materials scaling-up

The aim of this work package is to scale-up the production processes of the core materials required for the three CO₂ capture solutions addressed in the project. Within the present period the MMMs raw material production scaling-up has been completed. The specific achievements are detailed hereafter:

- Synthesis of MOFs (ZIF-8, MIL-140A-Br, ZIF-94 and NH₂-MIL-53) have been evaluated.
- All MOFs have been tested for separation performance in membranes ZIF-8 and ZIF-94 were chosen to go forward to scale-up.
- Several polymers have been tested using flat sheet membranes, Pebax 2533 was chosen for use in the prototype.
- Scale-up route for the polymer has been developed and 50 kg of polymer has been produced.
- Scale-up routes for ZIF-8 and ZIF-94 have been developed and 1-kilogram batches of both MOFs have been produced.

Investigation into the recycling of linkers and solvent in the MOF synthesis has been completed (Figure 5). The effect of different deprotonators as well as washing steps and drying procedure on the synthesis of MOF from the mother liquor was investigated. The morphology, thermal stability, crystallinity and surface area of the synthesized MOF were studied. This procedure enabled not only to obtain phase pure MOF but also to substantially decrease the amount of solvent used for washing making it a sustainable process.

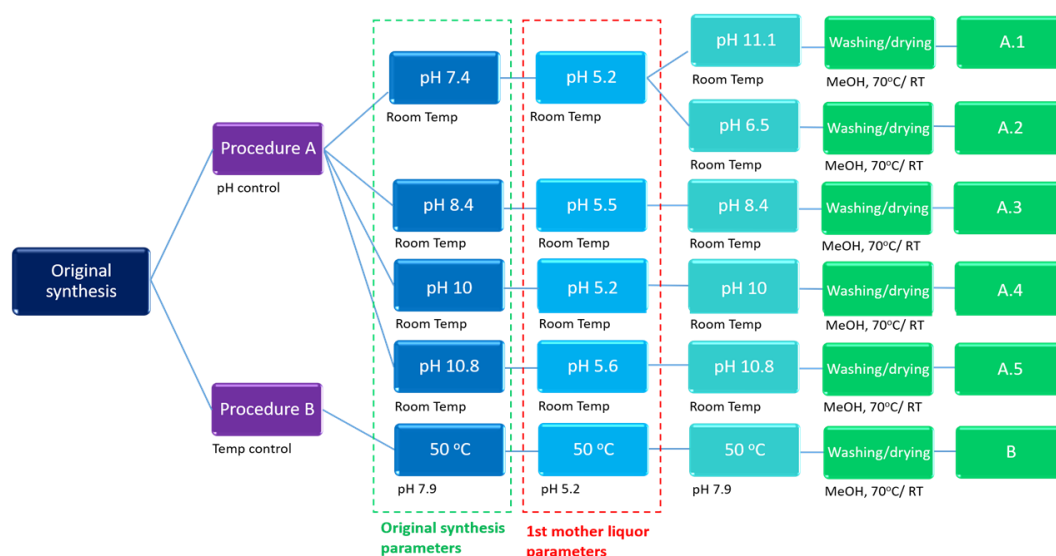


Figure 5. Solvent and ligand recycling in ZIF-94 synthesis.

Regarding the sorbent material production and scaling-up the following achievement have been reached:

- The sorbent has been optimised extensively specifically investigating precursor and water ratios, as well as heat treatment temperature.
- Several larger scale batches (20 kg) for evaluation have been produced and tested. An initial sorption capacity of 0.25 g-CO₂/g has been found to be optimum for the sorbent stability (Figure 6).
- Measurement of the Air Jet attrition Index to qualify/validate the mechanical performance of the sorbent is ongoing.
- Knowledge needed to produce the 200 kg batch for the prototype C test campaign has been generated, and manufacture will start once final material pre-validation is completed.

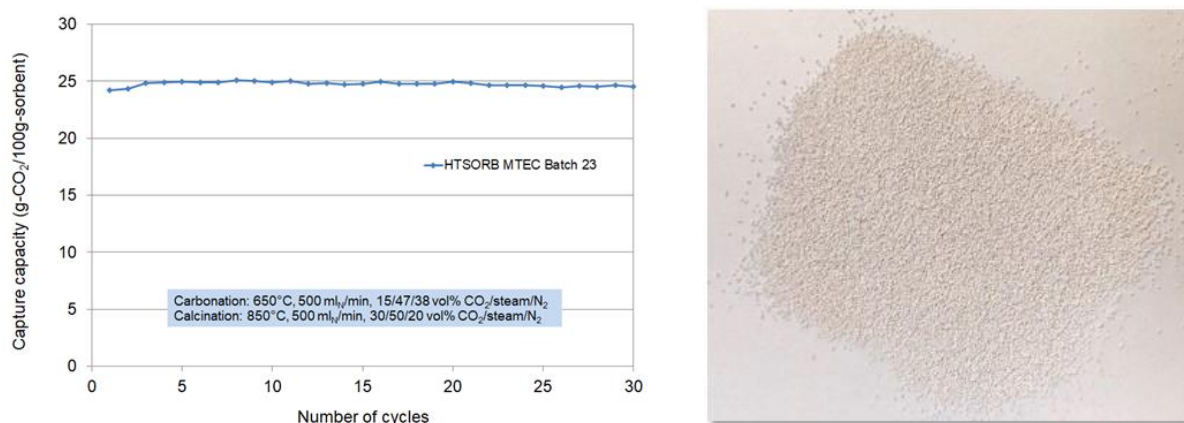


Figure 6. Sorption capacity (left) and HTSORB CO₂ sorbent (right).

Concerning the catalyst production and scaling-up the main activities and achievements are the following:

- Several catalyst formulations were synthesised and tested under standard reforming conditions.
- The catalyst C&CS #1005 mod 2 (Figure 7-left) was determined to be the best performing catalysts and was tested under small scale fixed bed sorption enhanced reforming conditions (Figure 7-right).
- C&CS #1005 mod 2 exhibiting a superior mechanical stability and catalytic activity than the initial C&CS #1005 has been scaled-up to 5 kg.
- Production of the 50 kg batch is underway.

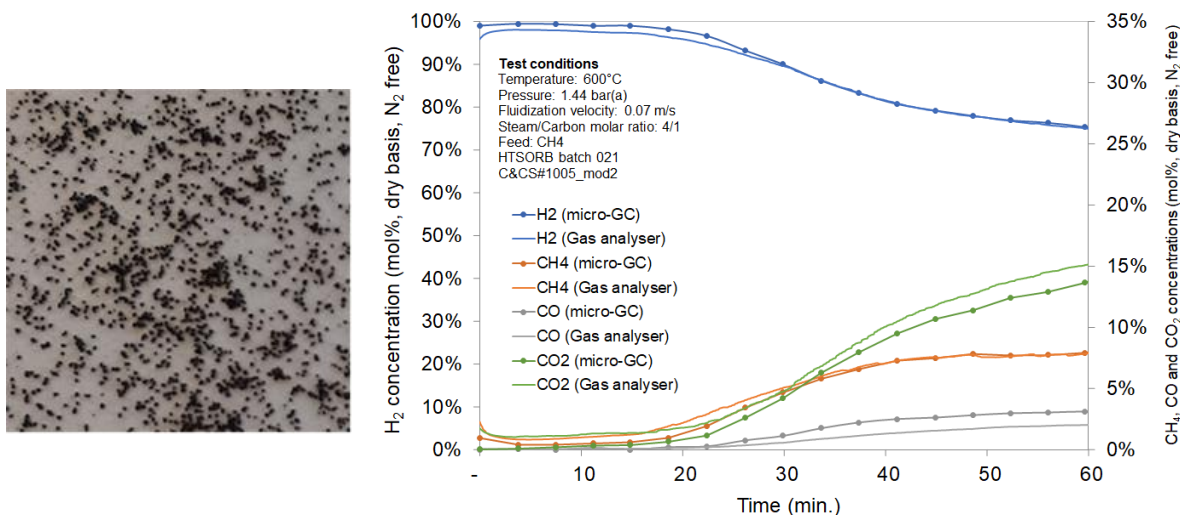


Figure 7. C&CS #1005 mod2 reforming catalyst in reduced form (left) and SER fluidized bed reactor test (at laboratory scale) in batch mode, using HTSORB sorbent mixed with catalyst #1005_mod2 (right).

Finally, large scale manufacturing of the MOF production has been also addressed:

- Industrial scale design for MOF production has been evaluated and proposed.
- Preliminary process flow diagrams for production of both MOFs has been completed including material balances and equipment sizing.
- Estimates of CAPEX and OPEX for MOF production plants have been calculated including production cost of the final materials.

Membranes production scaling-up

Along the present period, main activities have been focused on the optimization of membrane production parameters, the scaling up of the membranes for the prototypes and the optimization of recycling procedure of the critical membrane materials.

PBI based mixed matrix hollow fibre membrane spinning recipe has been optimized at lab scale. Significant improvements on hollow fibre processability have been achieved compared to the starting point spinning recipe (recipe developed in the previous reference M4CO2 project). On one hand, we managed to reach industrially relevant take up rate (25m/min), resulting in a substantial reduction on fibre diameter (from 470 μm to 270 μm). This will result in a significant improvement of the packing density of the prototype module (double surface area/volume ratio). Furthermore, PBI quantity required to produce one m^2 of hollow fibre has been halved. Spinning recipe has been transferred and the scaling up and production of the HF MMMs for the membrane module for the pre-combustion CO_2 capture prototype will be completed in the following weeks (Figure 8 - left).

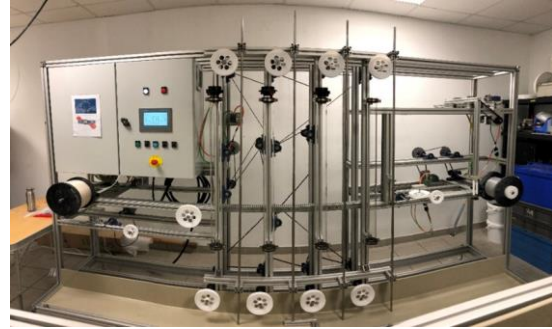


Figure 8. Pilot plant for scaling-up the production of HF MMMs: pre-combustion CO₂ capture (left) and post-combustion CO₂ capture (right).

Work is ongoing on the optimization of composite hollow fibre membrane preparation for the post-combustion CO₂ capture prototype (Figure 8 - right). Composite membrane is prepared using a continuous multi-layer coating device, where with one single production step a composite multi-layer dense membrane can be produced. The gutter layer coating has been optimized at large scale and optimization of selective layer coating is underway.

Membranes for the MA-SER prototype have been produced once the process parameter optimization for the fabrication of Pd-based membrane preparation was finished. 50 Pd-based membranes with an overall membranes surface area of 1.08 m² have been prepared. After successfully passing the quality control, the membranes have been delivered to TU Eindhoven where the integration in the reactor prototype C will be carried out.



Figure 9. Ceramic supported thin double skin Pd-based membranes prepared for the MA-SER prototype.

Activities related to the development of a specific and optimized procedure for the recycling of the metallic supports of the Pd-based membranes used in the MA-SER prototype has been continued as follows. On one hand, some adjustments have been performed to adapt the pilot plant to the optimized process and the pilot plant revamping has been completed. On the other hand, a wide laboratory experimental campaign has been carried out on Pd-based metallic membrane samples in order to optimize and define the recycling procedure. Furthermore, the lab experimental campaign on samples of Pd-based

ceramic supported membranes has begun to define their recycling procedure as well as the lab test on the recovery of Pd from leachate.

Pilot prototypes design, construction and testing

Within WP5 the partners are involved in the design and set-up of the three prototypes of MEMBER project plus the syngas cleaning system to be installed upstream the pre-combustion CO₂ capture prototype, Prototype A. The other prototype systems are: Prototype B, a post-combustion carbon capture system and Prototype C, the MA-SER system for H₂ production with integrated CO₂ capture.

The detailed engineering of the three prototypes and the syngas cleaning has been finalised. The specifications for all components have been made, and mechanical designs have been made for those components not commercially available. 3D designs of the prototypes have been made.

Using the specifications most of the components have been sourced and purchased. In the next phase construction will start.

Environmental and economic assessment

The first important step in this WP was the definition of the goal and scope of the Life Cycle Assessment (LCA) and Life Cycle Cost (LCC) analysis. The functional unit, to which all results of the study will refer is 1 kg of CO₂ captured and stored as well as for system C, the production of 1 m³ hydrogen.

To achieve all the goals set for the WP and to develop the required deliverables, a close and proactive collaboration with the project partners is needed. Quantis developed data collection files to have a clear baseline to model all the required key materials of the MEMBER systems.

The elaboration of the screening environmental LCA and LCC has been completed. This work aims at identifying the most significant areas of environmental impacts and costs with a focus on a cradle-to-grave framework. The work on this stage started at M13 with the development of data collection questionnaires which were sent to the partners. Based on the data, models were built which describe the three MEMBER technologies as well as the key materials. For elements for which no data was available from partners and for background data like electricity production or transport, the models rely on the existing datasets from data repositories. In the last months, the models were refined and transferred to an industrial level for the MEMBER lines, the reference cases and the key materials. The results, which were validated with the MEMBER partners, show the importance of the feedstock and therefore of the influence of carbon capture on the plant

efficiency in the reference systems, especially with regard to climate change impacts and use of resources.

The overall technical and economic assessment of MEMBER technologies as well as the identification of market potential in different sectors in which MEMBER configurations are suitable or can be adapted for specific applications, have already started and will be completed by the end of the project. This task is strictly linked to WP2, in which the technical evaluations for industrial application are carried out. Technical and economic viability for industrial application of the three MEMBER technologies is under investigation.

Dissemination and communication

Along this period the dissemination plan has been updated, including the consortium plans to make the MEMBER results known by the public. Internal and external communication tools have been updated (i.e. public website: <https://member-co2.com/>) as well as communication material (i.e. leaflet, poster, general presentation, newsletter) and MEMBER has been already disseminated in social media. Up to now 25 contributions to various conferences have been made, 3 articles in scientific journals published, one patent application filed and one thesis presented (<https://www.member-co2.com/content/publications>). A 1st public workshop on “Membrane processes for CO₂ capture”, organised by the MEMBER project, took place at TUE in January 15th, 2020. In addition, a second virtual workshop on “CO₂ Capture and Utilization” was organised at TUE by the MEMBER project jointly to other 11 projects. The meeting took place in February 16th-17th, 2021.

Highlights

MEMBER workshop 2020

Introduction to Membrane-based system for CO₂ capture Mid-January 2020, Eindhoven – The Netherlands

The workshop was a success with around 54 participants. The booklet with the public presentation of the workshop are available at the MEMBER website (<https://member-co2.com/>).

MEMBER joint workshop 2021

International workshop on CO₂ capture and utilization 16-17 February 2021, Eindhoven – The Netherlands

A second international workshop on “CO₂ capture and Utilization” organised by 11 EC funded projects in the field of CO₂ capture and utilization has taken place on February 16th-



17th 2021. The event was organised by our partner TUE with the support of the coordinators of the eleven EC projects (<https://www.iwccu.org/>).



The workshop was a success with around 256 participants from more than 26 different countries worldwide (Figure 10). Both the booklet with the presentations and the recorded presentations are now available online. You can download the booklet and find the playlist at the following link: <https://www.member-co2.com/content/workshops>.

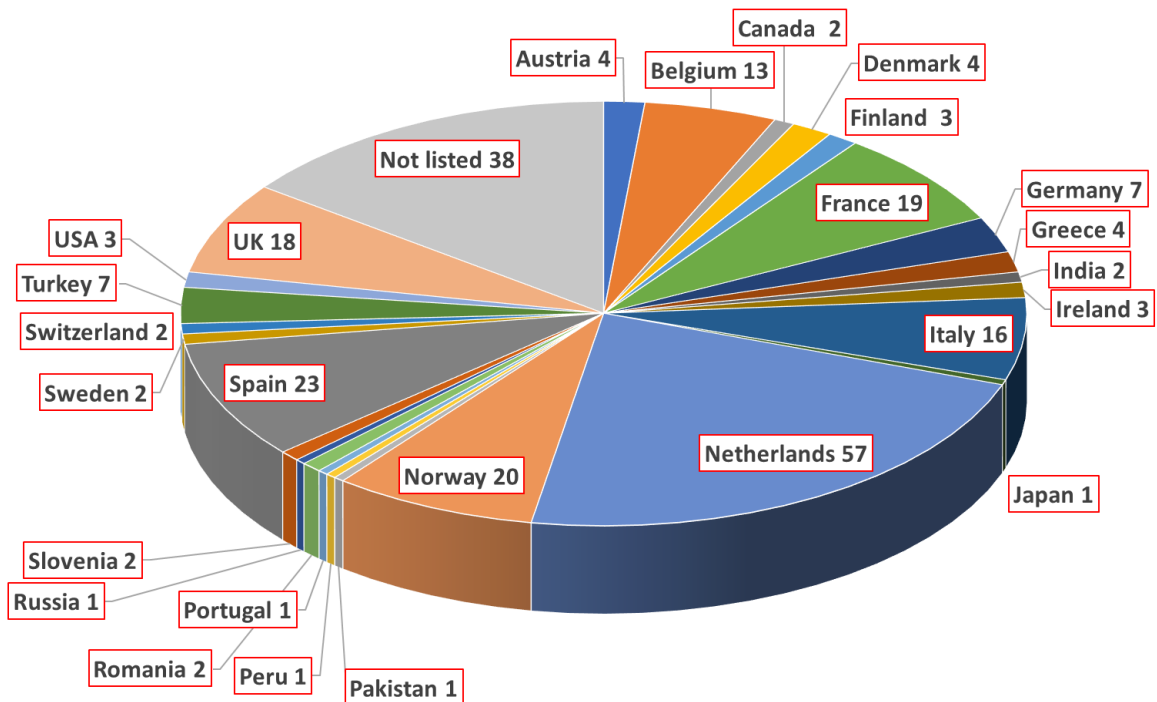


Figure 10. Participation by country.

MEMBER in figures:

- ↪ 17 partners (6 RES, 4 IND, 7 SME)
- ↪ 9 countries
- ↪ 9 596 541€ project (7 918 901€ EU funded)
- ↪ Start January 2018
- ↪ Duration: 48 months
- ↪ Key milestones:
 - ↪ February 2020 – three CO₂ capture concepts designed
 - ↪ December 2020 – prototypes ready for testing
 - ↪ December 2021 – demonstration of the prototypes in industrial relevant conditions at TRL 6

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More information on MEMBER (including a presentation of the project) is available at the project website: <https://member-co2.com/>

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Disclosure:

The present publication reflects only the author's views. The Commission is not responsible for any use that may be made of the information contained therein.

