31.01.2024

In the process industry, down-stream processes are the most resource and energy consuming industrial operation steps. Furthermore, the integration of new processes often requires a large portion of CAPEX and OPEX. To enhance the competitiveness of the European process industry and to contribute to Europe's goal of a clean and liveable environment, a broadly applicable concept for efficient integration of downstream operations in the overall process chain is highly desired.
The MACBETH consortium provides a ***breakthrough technology by combining catalytic synthesis reaction with the corresponding separation units*** *in a single highly efficient* ***C****atalytic* ***M****embrane* ***R****eactor (CMR). With this disruptive technology a* ***reduction of greenhouse gas emissions (GHG)*** *and an* ***increase in resource and energy efficiency*** *of large volume industrial processes can be achieved. The revolutionary new reactor design will guarantee substantially smaller and safer production plants and thus* ***reduce operational and investment costs****.*

MACBETH
Membranes And Catalysts Beyond Economic and Technological Hurdles

Topics

Consortium News

Consortium meeting in Rome

From 28 to 29 November 2023, MACBETH members met in Rome (Italy) for the 9th consortium meeting to discuss the status of the work and the next steps. Good progress was reported on all the flagship projects that bring catalytic membrane reactors one step closer to reality.

In addition to the official agenda part, MACBETH members had the opportunity to visit one of Rome's most important sights – the Trevi Fountain – on a guided tour. Following the tour, the participants enjoyed the specialities of Roman cuisine at a local restaurant.

The MACBETH consortium is looking forward to the next meeting scheduled for May in Istanbul.

Project Information

Project No.:
GA 869896

Call (ID) Identifier:
H2020-NMBP-SPIRE-2019

Topic:
CE-SPIRE-04-2019
Efficient integrated downstream processes (IA)

Project Duration:
54 (+6) months
Nov 2019 – Oct 2024

Project Budget:
20,7 M€

PROJECT PROGRESS - DEMO PLANTS

As outlined above, the MACBETH consortium provides a breakthrough technology combining catalytic synthesis reaction with the corresponding separation units in a single highly efficient catalytic membrane reactor (CMR). This innovative approach is applied to four crucial large-scale processes: Hydroformylation (HYFO), hydrogen production (H2), propane dehydrogenation (PDH) and bio catalytical oil cleavage (BOC). In the following, an update of the current status of each demonstrator for each of these four cases is provided.

HYFO - Hydroformylation

For the HYFO case the demo plant will be operated as a by-pass to the production plant at the Evonik Oxeno GmbH site in Marl (Germany). All main and auxiliary streams of the demonstrator are fed back into the production plant.

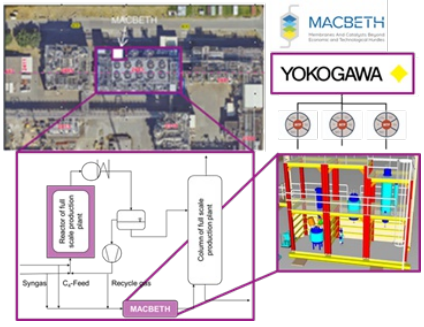

Figure 1: Integration of the MACBETH demo plant in by-pass to the production plant at the EPM site in Marl (Germany)

Since the demonstrator was designed in a modular fashion, it consists of three modules which were constructed external of the production plant (09 / 2022). The three modules are a catalyst coating unit, a fixed bed reactor unit for benchmarking purposes and the

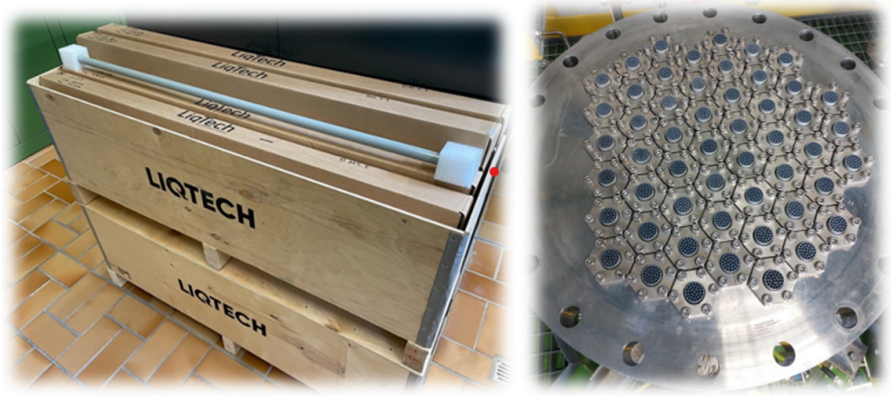

Figure 3: Placement of the membrane coated monoliths (left) in the MACBETH reactor (right)

MACBETH catalytic membrane reactor unit. A fourth module – a membrane unit – is intended to be implemented at a later stage within the downstream of the fixed bed reactor unit. After successful construction and cold start-up (06/2023), the modules were transported to the operating site (07/2023) and connected to the production plant (09/2023).

This procedure ensured minimal disruption to daily production routines.

Figure 2: Roadmap to implementation of the HyFo demo plant into by-pass of full scale production plant

At the production site the catalyst filling for both reactors took place. For the MACBETH reactor 52 membrane-coated monoliths were carefully placed inside the reactor ensuring that the membrane at the outside of each monolith was operational.

In a first stage the fixed bed reactor was started-up (11/2023) according to a procedure that was tested in Mini plant scale before in lab environment. In a second stage the MACBETH membrane reactor followed just before Christmas. Initial results indicate successful implementation of the membrane coated monoliths in the MACBETH reactor. Currently both reactors are running at a reduced capacity still carefully checking the behavior of the two reactors under various conditions and fixing smaller shortcomings. This implementation was only possible by a great collaboration of different teams

at Evonik including people from RD&I, Engineering, Process Technology, Technical Services and especially experts from the production plant.

The whole HYFO team from Evonik is now looking forward to the upcoming months of demonstration knowing that there will be challenges to be solved.

H2 - Hydrogen Production

H_2 demo plant in Spain

H2SITE has finalised the commissioning of the new reactor for the NG reformer. This new reactant feeding to the reactor has been modified to assure better performance (higher H_2 production and longer membrane stability). These modifications have been validated in an 8-hours automatic test without membranes. Up to now, a total of 250 hours of H2 production have been achieved.

The system now works in a fully automated mode, which means that the demonstrator can be operated with one single button. Once it is ready, the operator can start with the H_2 production phase. Besides, whenever is required to switch off the demonstrator, it is possible to single-press the button that ends the H_2 production phase. All these tasks can be performed either from the system screen (located next to the demonstrator) or remote-controlled from another place.

Next steps include the membrane integration to perform an 8-hours FAT test in the H2SITE facilities. After that, the reactor will be ready to obtain the CE-marking (which is only lasting for the last functionality test) and can be relocated from H2SITE to CNH2 with all required documentation. As soon as it gets here, commissioning at the new location will proceed until the start of the testing campaign, targeting more than 1000 hours of H_2 production.

The tasks to be carried out by CNH2 consist of preparing the facilities to host the MA-SR (membrane assisted steam reforming), in order to run it in safe conditions.

Preliminary works completed, such as the installation of a hydrogen compressor, civil works and the improvement of safety measures.

Before its arrival, the main ongoing work is the completion of the natural gas line to supply the MA-SR. Natural gas serves a double purpose, a part is burned to generate steam, to mix it with natural gas provided by the same line in presence of catalyst. This way, the reforming reaction occurs, producing hydrogen.

To feed the reactor, natural gas from the grid must be provided at high pressure, which is increased by a compressor.

Figure 5: The hydrogen compressor placed in its location at CNH2 (Spain)

The next step is the commissioning of the natural gas compressor to ensure that it will work as expected. This is a critical point of the system, because in case of failure it would not only avoid the MA-SR to run, but it would also be dangerous, due to the risk of gas leak, fire or explosion. At this point, the commissioning is under planification together with the company contracted to do it.

The final stage involves the connection and completion of the natural gas line, once the MA-SR arrives. After the completion, the line will be inspected and legalized to enable natural gas supply.

Once the installation and commissioning of the reforming are completed, the operation phase will start and the system will commence for an extended duration, in order to evaluate the behavior, efficiency and reliability of catalyst, membranes and the overall system.

Figure 4: Demo H2SITE(left) and instllation of the new reactor (right) (Spain)

H_2 Demo plant in France

To improve the production of pure H_2 from biogas or natural gas, MACBETH is developing, building and demonstrating a novel reactor concept integrating H_2 separation in-situ during the reforming reaction in a single vessel under industrially relevant conditions.

In the framework of the ATR demonstration line for H_2 production from biogas, Tecnalia has scaled-up the membrane production up to 125 palladium-based-double-skin-membranes onto a ceramic support developed by the partner RKV. These sealed membranes will be integrated into the pilot currently building by the partner ICI Caldaie.

ICI Caldaie is now finalizing the assembly of the ATR pilot. The Factory Acceptance Tests are forecasted to be organized in the Q1/2024.

In parallel, the Bio-Valo Methelec Biogas Plant located in Ennezat (France) has begun preparation works, coordinated by Engie, to be able to receive and connect the ATR pilot or demonstration.

The project aims to deliver the pilot at the end of the Q1 of 2024 and to initiate the commissioning and realize long term demonstration test campaign until October 2024, which is the end of Macbeth project with the objective to produce around 100 kg of hydrogen per day. Preparation works include the installation of an additional desulfurization unit to avoid the inlet of sulfur into the reactor, as palladium membranes are highly sensitive to sulfur compounds.

Figure 8: Demo Engie-Bio-Valo Methlec (France)

During the demonstration, the ATR pilot will be feed by strongly desulfurized raw biogas, which will used for the reaction to produce hydrogen on-site and by the boiler which produce the required heat for the reaction.

With the Catalytic Membrane Reactor developed in the framework of Macbeth Project, biogas is converted into H_2 at a much lower temperature compared with a conventional system, resulting in an increased of the overall process efficiency and a significant decrease in volumes and auxiliary heat management units.

The system's performance was in parallel investigated through a modelling work. The results showed an efficiency of the plant implementing the innovative MR of about 61%, about 17% higher than the benchmark technology. The direct CO_2 emissions and the cost of H_2 are about 28% and 30% lower, respectively than the benchmark technology.

PDH - Propane Dehydrogenation

During 2023, in order to face economical and technical issues, PDH partners had to modify the original program of demo plant realization. Therefore, instead of one demo plant at TRL7 at Engie premises in France, partners agreed to revamp three existing smaller plants to test PDH under different operating conditions. So, being the proposal approved by EC, the PDH process will be tested:

- at Tupras, Turkey at TRL6 (industrial environment) in non-integrated arrangement with membranes physically segregated from fixed catalytic bed,
- at TU/e, The Netherlands, at TRL5 (lab scale) in integrated arrangement with membranes inside

fluidized bed,

• at UNISA, Italy, at TRL5 in both arrangements with fixed catalytic bed. Moreover, the three plants will operate in different conditions in terms of pressure, temperature and space velocity, providing the opportunity to explore more extensively the behavior of catalyst and membranes in PDH process.

Revamping and procurement activities are in progress: in particular, membranes were sent from Tecnalia (Spain) and crossed Europe to reach the 3 demo plant sites. Our partners are now involved in the very delicate task of installing the membranes in their plants and conducting all the necessary tests for starting up their plants. It is anticipated that the three plants will be ready for the demonstration in Q2 of 2024.

In the meantime, KT and NextChem (a new partner enlarging the MACBETH family, who joined the project in March 2023) are in charge of design scale-up activities at an industrial scale, which will be finalized by end of MACBETH Project, October 2024.

Demonstrator: TUPRAS

As a demonstrator, TUPRAS started design and procurement activities in August 2023 for the revamping of its current plant to bring it to TRL-6 for demo operation. All the items are currently integrated to the system and TUPRAS demo plant is ready for operation. In the demo operation, there will be two reactors and one membrane separator which contains 7 membranes inside. The outlet stream of the first reactor will be directed to membrane separator for H_2 removal. To achieve effective H_2 removal, a driving force between the PDH atmosphere and permeate side is essential. Pressurizing the system could be an option for supplying driving force for H_2 removal.

However, increasing pressure in PDH leads to significant conversion decrease. Therefore, TUPRAS will perform the PDH demo operation at atmospheric pressure and apply vacuum to the inner side of the membranes to provide driving force for H2 removal. This will enable TUPRAS to carry out the demo operation with higher propane conversion and higher propylene yield per propane pass by using two scaled up catalysts, former is Pt (0.63% wt.) Sn (0.91% wt.), Sn/Pt: 1.45 on commercial Sasol MG70, the latter is Pt (0.35-0.5% wt.) with Sn/Pt ratio 1.40-1.50 on 93MG70 developed and shaped by TUPRAS.

Figure 7: Demo Tupras (Turkey)

Demonstrator: TU/e

The demonstrator operated at Eindhoven University of Technology in the Netherlands is of a much more experimental design, compared to the other two PDH demonstrators. The TU/e demonstrator investigates the option to integrate both the membranes and the catalyst in one single unit, achieving a maximum level of integration. This makes the design of the process much more compact, and it takes maximum advantage of the positive effects that the membrane and the catalyst have on each other. The main challenge lies in identifying process conditions that are compatible for both building blocks.

The TUE demonstrator is planned to start operation in January 2024.

Figure 8: Demo TU/e (Netherlands)

Demonstrator: UNISA

The plant at UNISA has been completed with:

- an additional mass spectrometer with magnetic sectors which will be installed in order to analyse the streams entering and exiting the membrane reactor. Coupled with the existing gas chromatograph could allow for more in depth understanding of both the catalyst and the membranes behavior;
- specifically designed ovens and reactors, both for the catalyst and the membranes, in order to realize a better controlled temperature profile.

Figure 9: Demo UNISA (Italy)

BOC - Bio Catalytical Oil Cleavage

The BOC case has some exciting developments brings forth. Firstly, the procurement process for all parts of the demonstrator plant has been finalized by the partners from Microinnova. Afterwards, construction of the pilot plant began and was completed on schedule in the fall of 2023.

The plant was then shipped to the labs of Enzymicals for a 5 month stop-over. Here, the pilot unit was fully assembled and tested under operating conditions for the very first time. As reported previously, initial trials showed encouraging results meeting targets for the main KPIs.

Currently, ongoing optimization experiments are being carried out allowing for a smooth transition from lab to an industrial environment at Solutex in Spain. Currently, further transportation of the pilot unit to Spain is being prepared. Additionally, the membrane part of the final demonstrator is ready for shipment and waiting for its

Figure 10: Installation Demo ENZY (Germany)

departure at the labs of VITO.

In the next future, in Spain at the industrial site of Solutex, both the enzymatic flow reactor and the membrane part will be integrated for the first time. Therefore, technicians from Enzymicals, VITO and Solutex will collaborate to successfully showcase the full potential of the omega-3 enrichment process developed in MACBETH.

All the involved partners are looking forward to this event with great optimism! Long-term stability tests will be performed and data will be generated to perform a life cycle analysis, to get a first estimation of the environmental impact of the novel process compared to the state-of-the-art technology.

In the meanwhile, fully in-house immobilized enzyme production has been achieved. From enzyme fermentation by Enzymicals to enzyme immobilization by Chiralvision; the provision of catalyst formulation could be secured within the team.

Figure 11: Demo ENZY (Germany)

EVENTS

Consortium Meetings

The 10th MACBETH Consortium Meeting will take place from the 14th to the 16th of May 2024 in Istanbul (Turkey). The meeting will be hosted by Türpas.

During case-wise workshops, recent results & technological progress will be discussed and presented in plenary sessions to the consortium. Additionally, communication, dissemination, and exploitation activities will be coordinated for the next project period.

The evening before the meeting, all project members will gather for get-together to foster social interaction.

The final consortium meeting is planned for the fall 2024 in Marl, Germany, hosted by Evonik.

Conference Participations

We have actively participated in conferences worldwide over the last few months. There are some highlights:

Annual Meeting of Reaction Engineering: Asem Al-Shaibani (FAU) participated in the Annual Meeting of Reaction Ingineering from May 15th to 17th, 2023, in Frankfurt, Germany with his presentation on Rh-catalyzed but-1-ene hydroformylation in a continuously operated gas-phase membrane reactor.

WCCE11: EVONIK took place in the 11th World Congress of Chemical Engineering from June 4th to 8th, 2023, in Buenos Aires, Argentina. Irin Wilson Panjikkaran delivered a presentation on "Modeling of an intensified reactor for hydroformylation of 1- butene with simultaneous catalytic gas phase reaction and membrane separation".

ICOM 2023: The MACBETH

consortium successfully partipicipated in the International Congress on Membranes and Membrane Processes from July 9th to 14th, 2023 in Chiba, Japan with two individual presentations.

ECCEAB23: The MACBETH consortium successfully partipicipated in the 14. European Congress of Chemical Engineering from September 17th to 21st, 2022, in Berlin, Germany with five individual presentations.

Europacat 2023: MACBETH was presented at Europacat 2023 from August 27th to September 1st, 2023 with two posters on MACBETH from DTU.

CYCSC2023: Irmak Su Okten (Tüpras) participated in CYCSC 2023 from 11th to 14th August in Novi Sad, Serbia, with her presentation about the Preparation of Pt based Hydrotalcite Derived Mg(Al)O Shaped Catalysts via Wet Impregnation for Propane Dehydrogenation Reaction.

CLASSY Industry Workshop: On October 16, 2023, David Liese (ENZY) presented on the topic "Recent results from the EU-funded project MACBETH: Biocatalytic omega-3 enrichment in fish oil in a packed bed flow reactor utilizing immobilized enzymes – From proof-of-concept to an industrial pilot plant in less than 4 years!". The workshop was held online.

Meet MACBETH

Are you interested to meet our partners and learn more about the project?

Upcoming Events:

March 06 – 08, 2024
ECHEC 2024
https://ehec.info/

June 10 – 14, 2024
ACHEMA
https://www.achema.de/en/

2nd half of 2024
Public demo site visits
https://macbeth-project.eu

Stay tuned! We will publish more information soon.

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www.twitter.com/MACBETH_H2020 | www.linkedin.com/company/macbeth-h2020

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