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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

Last year, our project held two extraordinary consortium meetings. The first meeting took place in May 2024 in Istanbul, Turkey, organized by the consortium member Tüpraş. Participants had the opportunity not only to work but also to explore the city and visit the Tüpraş refinery.

The final consortium meeting was hosted by Evonik in December 2024 in Germany. In addition the presentations, participants visited the MACBETH demonstration plant at the Evonik facility. The meeting also included a guided tour of the Borussia Dortmund football stadium and the Zeche Zollverein industrial museum.

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 – Jan 2025

Project Budget:
20,7 M€

PROJECT PROGRESS - DEMO PLANTS

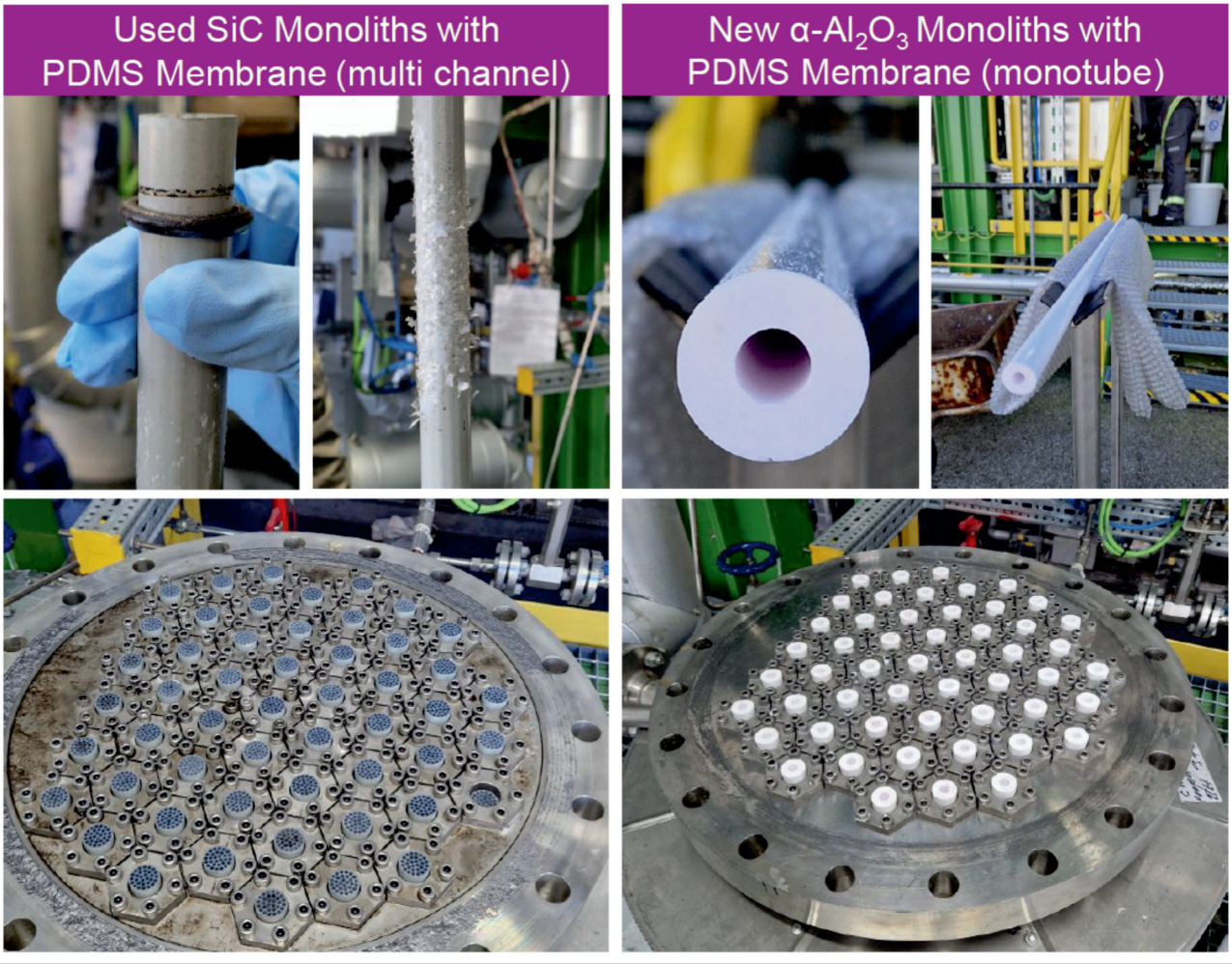
As outlined above, the MACBETH consortium provides a breakthrough technology combining catalytic synthesis reaction with the corresponding separation units in a single highly efficient catalytic membrane reactor (CMR). This innovative approach is applied to four crucial large-scale processes: Hydroformylation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

HYFO - Hydroformylation

The MACBETH demonstrator for the HyFo case was successfully operated for more than 5,000 hours in bypass to the 2-PH world scale production plant at the OXENO site in Marl, Germany. During that campaign, the reactor was operated by the plant operators. This first run allowed valuable insights into the operation of that new technology in the field (e.g. stability) triggering already further optimization of the technology.

The first campaign was finished in July 2024 during the annual revision of the world scale production plant and the catalytic monoliths were discharged from the reactor for further analysis. In the same period the MACBETH demonstration reactor was reloaded with optimized monoliths from RAUSCHERT replacing the former SiC monoliths.

The reactor is now being operated in a second demonstration campaign that started at the end of November 2024 and will last untill the end of the project. The focus of that campaign is to test the new support material and geometry, some optimized start-up procedures and the refined membrane coating.



H2 - Hydrogen Production

H_2 Demo Plant in Spain

The test campaign is finally approaching its conclusion. Each day, as usual, the MA-SR system is activated, producing high-purity hydrogen for several hours after warming up. The daily cycle of warming up, production, and cooling down presents a challenge to the system's reliability, yet it operates without any loss of performance.

In the final stage of the campaign, the primary challenge is the extremely cold outdoor weather, characterized by sub-zero temperatures, rain, wind, and ice. This harsh conditions extend the time required to reach the operating temperature compared to autumn conditions. Nevertheless, the results demonstrate that thermal stress is not a critical issue for natural gas reforming and its membranes.

To date, the system has completed over 400 hours of operation, including 126 hours of hydrogen production. The next step, following the final shutdown, will involve decommissioning the system in collaboration with H2Site to facilitate its return home.

H2 demo at CNH2 (Spain)

H_2 Demo Plant in Italy

The prototype of ICI (biogas autothermal reforming) was supposed to be tested in France by ENGIE. Unfortunately, despite a very long process, the authorities in France were unable to grant the permits to operate the prototype (not due to technical problems with the reactor, but rather bureaucratic issues). It was therefore decided to prepare the demo site at ICI in Italy and test the prototype directly there, these tests are in progress now and will continue beyond the project.

The membrane reactor system for biogas reforming remains an interesting business opportunity for small-scale and decentralized production of H2.

H2 demo at ICI in Italy

PDH - Propane Dehydrogenation

Demonstrator: TUPRAS (TRL 6)

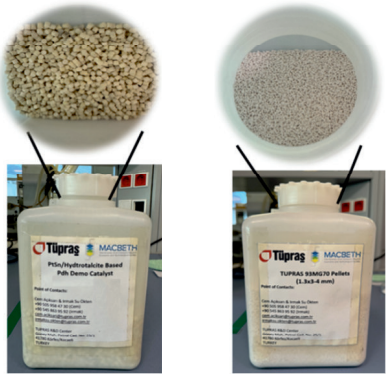
The PDH demonstration phase started at TUPRAS X2L PDH reactor system after the revamp of the unit. After integrating higher capacity MFCs and Coriolis, new furnace, electronic rotameter and other required items to the unit, TUPRAS PDH reactor system was ready for demonstration. Within the scope of demo operation, two reactors configuration with one membrane separator in between will be used. With seven membranes installed in the membrane separator, H2 will be effectively separated from the first reactor outlet stream and separated gas will be fed to the second reactor while separated H2 will be sent to the vacuum line. A vacuum of up to 100 mbar will be applied to the inner sides of the membrane to provide pressure difference between permeate and retentate sides to drive the separation. PDH demonstration will be conducted at 550 ◦C, atmospheric pressure and WHSV: 4 with two reactors loaded 413 g catalyst per reactor. Before starting the demo operation, TUPRAS has completed the preparation phase of the demonstration. Rather than immediatly starting the demonstration with 1,500g/h propane flow and integration and seven membranes, TUPRAS installed only one membrane and conducted some start up

Propane dehydrogenation unit

Membrane separator for hydrogen removal

studies. After finalizing the preparation phase, TUPRAS will commece PDH demonstration for 500 h.

Furthermore, both commercial and homemade extruded supports were Pt-Sn impregnated according to the formulation developed by Unisa and scaled up to approximately 1 kg for use in the 500-hour demo operation. The upcoming demo operation is designed to investigate the impact of membrane-integrated reactor technology on increasing propylene yield and to provide significant insights for the widespread adoption of this innovative technology in the industry.

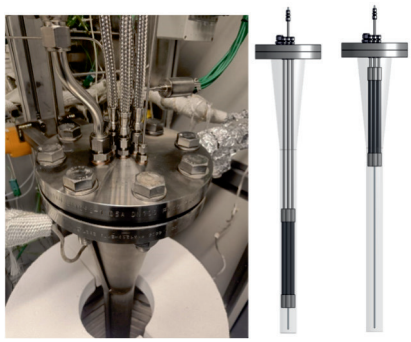


Scaled-up commercial supports (left); Scaled-up extruded supports (right)

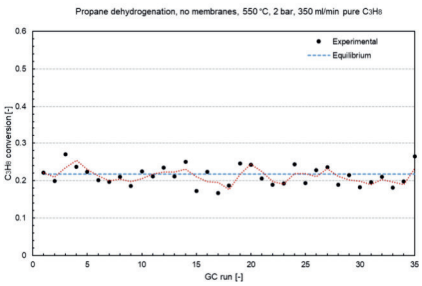
Demonstrator: TU/e (TRL 5)

At TU/e, the membrane-assisted propane dehydrogenation demonstration will be performed in a stainless-steel tubular membrane reactor (internal diameter = 52.51 mm, length excluding freeboard = 400 mm) accommodating four Pd-Ag membranes, which can be skid upwards through the reactor's freeboard to facilitate regeneration. The membranes will be immersed in a fluidized bed of catalyst (300 g, developed in-house, Pt Sn/HTC MG70, UNISA).

The results highlighted a umf of ~0.003 m/s at 20°C, which decreased to ~0.0012 m/s at 500°C. This led to a feed selection of ~2x umf, corresponding to ~350 ml/min of C3H8, accounting for the additional production of H2 given by the PDH reaction.



Fluidized bed membrane reactor for PDH demonstration at TU/e (left) and its internal design for membrane regeneration (right).

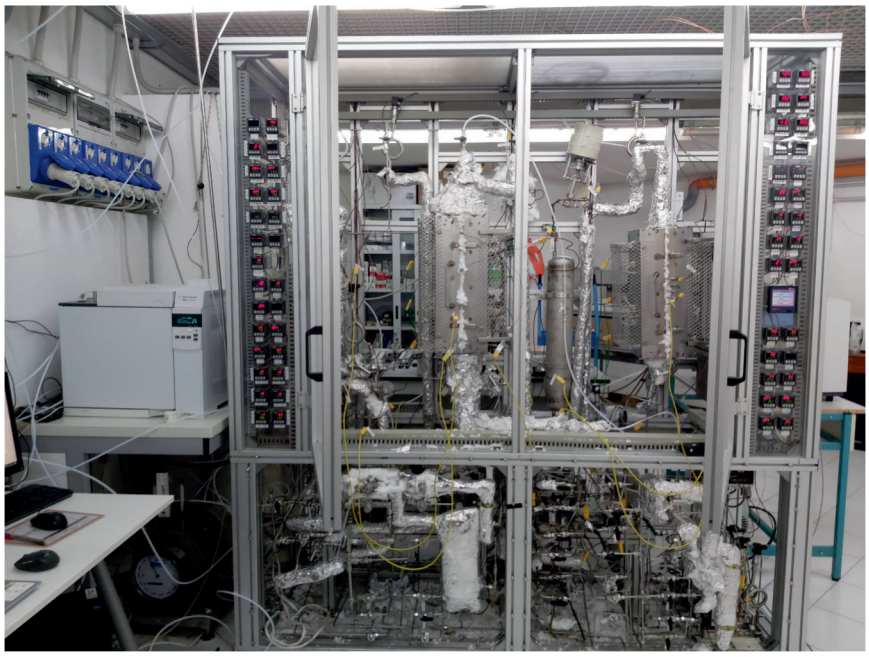


Preliminary C3H8 conversion at 550°C, 2bar, 2x umf per GC injection. The test was performed as preliminary factory acceptance test to evaluate the acceptability of the experimental error.

Demonstrator: UNISA

Beginning in December 2024, the UNISA demonstrator commenced operation, utilizing both the PDH catalytic reactor and the Membrane Separator within the framework of the so-called "hybrid architecture".

The activities have resumed and are scheduled to continue until the end of January, in alignment with the original timeline. To date, over 50 hours of operation have been recorded, with results confirming the excellent performance of both the catalyst and the membrane. Specifically, the catalyst demonstrated a propane conversion rate very close to the equilibrium limit, alongside propylene selectivity values exceeding the project target of 95%. Moreover, the catalyst exhibited remarkable stability and an exceptionally low tendency to coke formation compared to conventional PDH reactors.



Demo UNISA (Italy)

BOC - Bio Catalytical Oil Cleavage

After extensive testing of the individual pilot units, the complete BOC pilot plant was successfully assembled at the Solutex laboratories in Spain. Since then, we have accumulated over 2000 hours of demonstration time. Over 4,000 litres of crude fish oil have been processed during the trials. The demonstrator runs 24/7 at a flow rate of 1.4 L/h. Various operating parameters have been tested at TRL 6 and the entire process is already being continuously optimised.

Initial analytical results are very promising. Starting with an omega-3 concentration of ~30% in the raw material, it can be shown that the value has been increased to over 50% in the final product. However, there is still room for improvement:

Optimised catalysts with improved selectivity profiles have now been developed and tested on a smaller scale. Promising candidates have been identified and produced on a large scale for testing under operating conditions. These tests will be completed in the first quarter of 2025 to conclude the demonstration phase of the BOC case.

Fully assembled BOC pilot plant in the labs of Solutex.

EVENTS

Consortium Meetings

The final review MACBETH Meeting will take place on February, 12 2025 at EVONIK in Marl (Germany).

The final case-wise and work-package results as well as the technological progress, will be discussed and presented to the project officer and monitor.

Conference Participations

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

EHEC 2024: Mathilde Jégoux (ENGIE) participated in the European Hydrogen Energy Conference from March 6th to 8th, 2024, in Bilbao, Spain with her talk on MACBETH - Membranes and Catalysts Beyond Economic and Technological Hurdles: Improved membraned reactor for a more efficient H2 production.

18th International Congress on Catalysis: DTU took place in the 18th Internatinal Congress on Cataylsis from July 14th to 19th, 2024, in Lyon, France. Anders Riisager delivered a presentation on "MacBeth – Progress on the implementation of more sustainable hydroformylation by supported liquid phase (SLP) catalysis" and a poster on "Characterization and structure-activity relationship of a SLP system for more sustainable gas phase hydroformylation of 1-butene" as well.

Euromembrane 2024: The MACBETH consortium successfully participated in the international conference focused on membrane scienceand technology from September 8th to 9th, 2024 in Prague, Czech Republic with a presentation from ENGIE and a poster from ICI.

BIOCAT2024: Dr. Rob Schoevaart gave a lecture on "Hundred years of enzyme immobilization, what's next?" at the BIOCAT2024 on August 26th, 2024, in Hamburg, Germany.

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