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In the process industry, down-stream processes are the most resource and energy consuming industrial operation steps. Moreover, 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 Catalytic Membrane Reactor (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.



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# **Consortium News**

EC president von der Leyen praises MACBETH predecessor ROMEO

On November 10th 2021, European Commission president Ursula von der Leyen held a key note speech at the "Evonik Perspektiven" event. As part of her speech, she spoke about the Chemical industry's become strive to more sustainable and about the key contribution projects predecessor ROMEO (the project of MACBETH) can make to reduce the industry's carbon footprint. About the ROMEO project and its significance, von der Leyen said the following (English translation):

"Those who want to see the future of the chemical industry, have to visit you in the Ruhr area. I am thinking, for example, of a project with the beautiful name "ROMEO". [...]



Many major industrialized and regions have nations issued ambitious climate targets. Examples like ROMEO show that the chemical industry is already on the way to more sustainability. Chemicals are a crucial building block for solar systems plants or for the rotor blades of wind turbines. But the chemical industry is also a

# **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€



pioneer when it comes to reducing emissions of greenhouse gases. Since 1990, it has cut its emissions by more than half. And projects like the ROMEO reactor show how consistently you continue to speed up. I can only assure you: Europe will give you a tailwind along the way."

#### Project Videos

Over the last months, we have been constantly working on several project videos to visualize and explain ambitious MACBETH goals. In September 2021 we proudly released our first MACBETH project video presenting the breakthrough technology the process industry, our highly efficient catalytic membrane reactor (CMR) which combines the catalytic synthesis step and the separation step using a tailor-made membrane achieve the goal of a climateneutral tomorrow.



**MACBETH** partner Evonik released another video in January 2022 explaining MACBETHs benefit in revolutionizing large-scale reactions such as hydroformylation by making them more energy efficient and thus reducing emissions. The process already works in the lab and now the process is being tested in a demonstration plant industrial conditions at the Marl Chemical Park.



An important part of our MACBETH project is education. For this purpose, our first educational video on how Pdmetallic supported hased membranes are developed has been prepared and released in April 2022 by MACBETH partner Eindhoven University Technology. Within the video, individual steps are explained and visualized. We are already curious what other videos will be created in the future!

TU/e DEPARTMENT OF CHEMICAL ENGINEERING AND CHEMISTRY

Sustainable Process Engineering



# Otto-Roelen-Medal for Prof. Dr. Robert Franke

Our project coordinator Prof. Dr. Robert Franke received the Otto-Roelen-Medal 2022. With this award his pioneering contributions in the field of catalytic carbonylation, especially hydroformylation, from its theoretical foundations to its industrial implementation is recognised.



# PROJECT PROGRESS

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 mem-

brane reactor (CMR). In the project, this technology is applied to four highly relevant large-scale processes: Hydroformylation (HYFO), hydrogen production (H2), propane dehydrogenation (PDH) and bio

catalytical oil cleavage (BOC). In the following, an update on the project progress achieved for each of these four cases is provided.

# HYFO - Hydroformylation

In the Hydroformylation (HYFO) case, a catalytic membrane reactor is applied to the large volume production process of valeraldehyde. In the laboratories of Evonik's process technology department in Marl

the technology has been developed and evaluated for more than 5 years. Although a lot of experience has been gained on laboratory scale, an evaluation under industrial conditions is essential. For this

reason, the membrane reactor will be integrated into a side stream of the so-called Oxo plant at the Marl Chemical Park. Recently, an important milestone on the way to industrial application has been



reached. One important characteristic of the technology is the in-situ coating of the carrier with catalyst solution. On lab scale the solvent Dichloromethane has been originally used very а but powerful solvent also cancerogenic substance. "Safety is our top priority, therefor it was essential to find alternative solvent to transfer the heterogenized hydroformylation to industrial environment." says Prof. Dr. Franke head of

hydroformylation research at Evonik.

In the past years possible alternative solvents have been identified and tested with the help of the project partners. Dissolving all the catalyst components is quiet challenging task. That is why the project team was thrilled to have found, that Pentanal can be used as solvent. It can be applied according to the known procedure (Patent#: EP3632888A1), so no changes to the technology are

necessary. Furthermore, since it is the product of the target reaction, it is already present in the process, which simplifies the process design and increases economic viability.

With fulfillment of this vital prerequisite, the HYFO-case is now good on track to a successful integration of the demonstrator in the production plant. The basic design phase is currently conducted with an anticipated start-up in spring 2023.

### H2 - Hydrogen Production

In the Hydrogen line, two systems are being developed for pure hydrogen production through membrane reactors. One prototype is based on steam reforming, for this part H2Site (Spain) is responsible for the development of the which prototype, is well advanced and is now being finalized and will soon be ready for initial tests. The second based prototype is on autothermal reformina of biogas and ICI Caldaie (Italy) is finalizing the acquisition of the components before installing the system. For the reactor of ICI Caldaie, the membranes are ready as produced by Tecnalia. In the meantime, catalyst formulation is being optimized by JM with the modelling support of POLIMI and TUE.

In the meantime, the sites for the testing of the two systems are being prepared, with the one of CNH2 (in Spain) ready to receive the first reactor from H2Site.

Finally, new membranes are being developed in the hydrogen line, based on cheap metallic supports that are being modified to be used as Pd supports. TUE is carrying out membrane preparation at labscale, with the support of Tecnalia (see the video in the previous section).

The Hydrogen line is well in track with the plan for testing, despite the COVID crisys and the international situation makes even more difficult to receive the parts for completing the systems. Several results of this line will be presented as oral presentations in Japan at the International Conference on Catalysis in Membrane Reactors in August.

# PDH - Propane Dehydrogenation

The PDH case is in a very important phase of the project: the detailed engineering (KT in charge) of the prototype is and as strong on collaboration with ENGIE and ICI has been put in place to better define the final prototype architecture and layout solution for installation at ENGIE premises.

In final definition of the prototype architecture and operating conditions, the input received from UNISA, relevant to catalyst operation, from

TUPRAS for catalyst scale up matter and from TECNALIA/TUE, for membrane optimal operating conditions, have been of outmost importance for the achievement of such goal.

Parallel to the modeling (POLIMI/UNIBS) and design activities (KT), the Business Plan and Exploitation activities are in progress; following the activities on the Value Chain Stakeholder and market analysis carried out in the second year of the project, the

focus is now currently on the definition of exploitation plans with the aim of identifying the commercial strategy for the novel PDH technology deployment.

meantime, the activities for the LCA are also in progress with the collection of general information on the benchmark process and on novel PDH process to outline for the framework data collection and the impact assessment methodology.



### BOC - Bio Catalytical Oil Cleavage

The Biocatalytic Oil Cleavage (BOC) case has achieved two important milestones of the project within the last couple of months.

The partners ChiralVision from the Netherlands and the VITO institute from Belgium were successful in showcasing proof of concept of the combining a biocatalyst and a membrane. Α ceramic membrane was modified by growing polymer chains from the surface of the membrane. These polymer chains contained functional groups that could be used to immobilize an enzyme to the membrane surface. First tests showed promising conversion for the reaction of interest. This marks a major milestone in the development of a joint catalytic membrane reactor unit in the

field of biotechnology.

Another huge progress was made in the development of the membrane part of the process. After screening and parameter optimization principle of the membrane separation part was put to the first test with an actual reaction mixture, which was obtained from experiments in the lab plant at Microinnova in Austria. It turned out to be yet another success story. The final product samples were analyzed in the laboratories of Enzymicals. We were delighted to see, that the membrane performed better than expected. Thus, increasing the overall selectivity of the process.

Further progress was made on the engineering part of the project. The user requirements for the pilot plant are almost finalized and the detailed engineering can start soon. Next tests include parameter optimization first in lab then in pilot scale. Also, long-term stability tests of the process as catalyst well as lifetime investigation will be performed in the upcoming months. In the first half of 2023 we expect to perform the first trials in pilot scale. In the meantime, POLIMI/UNIBS were able to establish a suitable model for the enzymatic reaction part of the process.



## **EVENTS**

#### MACBETH Webinar

The MACBETH consortium successfully held its first "Membrane webinar on Reactors: one step forward towards industrial implementation" on Friday 5th of November 2021.



After the case presentations our 55 participants had the chance to ask their questions and lively discussions followed. The recording of the webinar is available on the MACBETH YouTube Channel.

#### Consortium Meetings

The virtual 5th MACBETH Consortium Meeting was held

from 27th to 29th of October 2021 to discuss the outcomes and results of the recent progress. All partners contributed to this great meeting and put a lot of efforts to bring MACBETHs breakthrough catalytic membrane reactor technology forward despite difficult working conditions due to the pandemic.



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

period. For socializing, virtual fireplace discussions in the evening of day 1 enabled a gettogether of project members.

All project members are already looking forward to the next "offline" project meeting which is scheduled from 18th to 20th of May 2022 in Geesthacht, Germany. We are pretty excited to meet in person after such a long time!

### Meeting of the External Advisory Board

On Friday, 28th of January 2022 we had our External Advisory Board (EAB) Meeting. Due to the ongoing situation, the meeting was held virtually. Nevertheless, we had inspiring presentations of our four cases und got fruitful input from our Advisory Board. Thanks a lot to our team of experts for the inspiring questions and the useful input and discussions!



#### Conference Participations

We have been present in (virtual) conferences all over Europe in the last months. Some of the conferences we attended are presented in the following.

**ECCE 2021**: The MACBETH successfully parconsortium ticipated the European in Conference Chemical on Engineering during September 20th to 24th 2021. MACBETH was represented by a keynote lecture about the general idea of MACBETH followed by 9 presentations individual and posters of the four show cases.

CEHC-2: On March 29. - 31. 2022, DTU participated in the Cutting-Edge Homogeneous Catalysis in Leipzig, Germany. Leonhard Schill showed attendees the progress moving gas phase hydroformylation towards industrial application which was made possible by catalyst phase optimization and the use of a membrane reactor.

#### **Announcements**

ICCMR-15: The 15th International Conference on Catalysis in Membrane Reactors (ICCMR) will take place from 31. July – 04. August 2022 in Tokyo, Japan hybrid style (faceto-face and online). The MACBETH consortium will participate with four contributions.



**ACHEMA 2022:** The world's leading trade show for the process industries takes place from August 22 – 26, 2022 in Frankfurt, Germany. MACBETH will be present here with a joint presentation of our partners.



### Meet MACBETH

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

### **Upcoming Events:**

May 18 - 20, 2022

6th Consortium Meeting

June 06 - 08, 2022

19th Nordic Symposium on Catalysis

https://19nsc.fi/index.html

#### July 31 - August 04, 2022

15th International conference on catalysis in membrane reactors (ICCMR-15) http://iccmr15.org/

August 22 - 26, 2022

ACHEMA 2022

https://www.achema.de/de/





















































