Final report

1. Project details

Project title	Highly Flexible Energy Production by Oxy-Fired Biomass Gasification (HighFlex)
File no.	64018-0028 (before 12403 under Forskel/PSO)
Name of the funding scheme	EUDP (originally funded under Forskel PSO)
Project managing company / institution	Technical University of Denmark (DTU), Department of Energy Storage and Conversion (DTU Energy)
CVR number (central business register)	30060946
Project partners	DTU (Department of Energy Conversion and Storage (DTU Energy) and the Department of Chemical and Biochemical Engineering (DTU Chemical Engineering)), CoorsTek Membrane Sciences AS, Evida (former HMN Naturgas I/S), Danish Fluid Bed Technology Aps
Submission date	26 February 2021

2. Summary

The highly efficient use of biomass to generate electricity, heat, synthetic natural gas and transport fuel will be important for a future Danish energy system based on 100 % renewables. The *HighFlex* project contributed to this vision by demonstrating novel technical solutions that allow using the available biomass more efficiently with high flexibility.

The underlying idea was to operate biomass gasifiers with integrated *oxygen transport membranes* (OTMs). Using oxygen instead of air in the gasification process triples the heating value of the produced syngas and increases the gas purity. This will allow i) the use of highly efficient gas turbines for electricity generation, and ii) upgrading the syngas to synthetic natural gas for storage. OTM, the core technology explored in *Highflex*, is a promising alternative to produce oxygen for high temperature processes with lower Capex and Opex compared to conventional technology for oxygen production.

The direct outcomes of this project are i) a robust manufacturing route for the ceramic OTMs including an analysis for up-scaling, ii) a proof-of-concept operation of oxygen membranes in a gasifier and pyrolysis unit, and iii) an analysis with recommendations on how this technology can be integrated into the future Danish energy system.

The results from *Highflex* will be directly used in a recently granted large EU project (FlexSNG, H2020, coordinated by VTT, Finland). In this project, DTU will continue the work started in *Highflex* by building a larger proof-of-concept module based on *OTMs* with several (industrial) stakeholders. The pyrolysis technology used in *Highflex* is currently scaled up to MW scale by the Danish company Stiesdal A/S; here OTMs are among the possible alternatives discussed for oxygen delivery.

Dansk resume

Effektiv anvendelse af biomasse til produktion af elektricitet, varme, syntetisk naturgas og transportbrændstof vil være vigtig for et fremtidigt dansk energisystem baseret på 100% vedvarende energi. *HighFlex*-projektet bidrog til denne vision ved at demonstrere nye tekniske løsninger, der gør det muligt at bruge den tilgængelige biomasse mere effektivt med høj fleksibilitet.

Den bagvedliggende idé var at drive biomasseforgassere med integrerede ilttransportmembraner (OTM'er). Brug af ilt i stedet for luft i forgasningsprocessen tredobler opvarmningsværdien af den producerede syngas og øger gasens renhed. Dette giver mulighed for i) brug af højeffektive gasturbiner til elproduktion, og ii) opgradering af syngas til syntetisk naturgas til opbevaring. OTM, kerneteknologien, der er udforsket i *HighFlex*, er et lovende alternativ til at producere ilt til højtemperaturprocesser med lavere Capex og Opex sammenlignet med konventionel teknologi til iltproduktion.

De direkte resultater af dette projekt er i) en robust fremstillingsvej for de keramiske OTM'er, inklusive en analyse til at skalere deres produktion op, ii) en proof-of-concept-drift af iltmembraner i en forgasser- og pyrolyseenhed, og iii) en analyse med anbefalinger om, hvordan denne teknologi kan integreres i det fremtidige danske energisystem.

Resultaterne fra *HighFlex* vil blive brugt direkte i et nyligt tildelt stort EU-projekt (FlexSNG, H2020, koordineret af VTT, Finland). I dette projekt vil DTU fortsætte det startede arbejde i *HighFlex* ved at opbygge et større proof-of-concept-modul baseret på OTM'er med flere (industrielle) interessenter. Pyrolyseteknologien anvendt i *HighFlex* skaleres i øjeblikket op til MW-skala af det danske firma Stiesdal A / S; her er OTM'er blandt de mulige alternativer, der diskuteres til iltlevering.

3. Project objectives

Objective of the project

The long-term vison behind the *HighFlex* project is to establish oxygen blown biomass gasification as highly efficient method to poly-generate electricity, heat, synthetic natural gas and transport fuel for the Danish energy sector from waste biomass. To support the development of a demonstration plant combining state-of-the art gasifier technology and highly efficient oxygen transport membranes (OTMs) for oxygen production in the future, the following project objectives were defined:

i) OTMs with high performance, robustness and long-term stability will be developed and demonstrated

ii) An analysis of how the production of these OTMs can be scaled up to relevant industrial scale

iii) Evaluation of the OTMs for producer gas upgrade via partial oxidation of tars

iv) Identification of the most optimal way to integrate the membranes

v) Recommendations and guidelines for a 100 kW demonstration oxygen blown gasifier supplied 100 % from OTMs

vi) An analysis how oxygen blown biomass gasification can be integrated in the future Danish energy system

All of the project objectives listed above have been reached.

Background on developed and demonstrated Technology

In the following section, the relevant background of the underlying technologies, biomass gasification/pyrolysis and oxygen transport membranes, are summarized.

Biomass Gasification and Pyrolysis:

Thermochemical conversion processes (combustion, gasification and pyrolysis) are widely used to recover energy from biomass. Especially pyrolysis and gasification are of interest, as both platforms offer high feed and product flexibility, providing the possibility to convert many different biogenic feedstocks into a wide variety of products such as heat, electricity, synthetic natural gas (SNG), chemicals, transport fuels and high value ash and char products.

Pyrolysis is a thermal decomposition process of carbonaceous materials in the absence of air/oxygen. The cracking of chemical bonds leads to the formation of molecules with a lower molecular weight. Different product fractions are obtained: a solid (char), a liquid/condensed (tars) and a non-condensable gaseous fraction. Depending on the heating rate and solid residence time, biomass pyrolysis can be divided into three main types: slow pyrolysis, fast pyrolysis and flash pyrolysis.

The gasification platform adds to the pyrolysis a char conversion process where carbon in the char reacts with a gasification agent such as steam or carbon dioxide at elevated temperatures. Thus, gasification is a partial thermal oxidation of carbon-rich materials yielding a non-condensable gas product (CO_2, CO, H_2, H_2O and other gaseous hydrocarbons) and smaller quantities of by-products such as char, ash, and condensable fractions including water and tars. Biomass gasification platforms are most commonly designed for conversion of wood in the form of wood pellets, wood chips or waste wood. However, it is possible to extend the potential range of organic material fractions converted in thermal gasification to cover various other organic resources. This includes agricultural, municipal and industrial by-products and residues such as cereal straw, fiber residues, sewage sludge and many further municipal and industrial organic waste fractions.

The gasifying agent for biomass gasification at moderate temperatures is usually either air, steam, pure oxygen or their combination. Air is widely used as an oxidant for the biomass gasification, with equivalence ratios of 0.2–0.3 (O_2 supplied/O_2 required for stoichiometric combustion), because of its low-cost availability. However, the producer gas from air-blown biomass gasification contains around 30–50 vol% N_2, has a lower heating value (4–7 MJ/Nm^3) and is therefore mostly used for heat and power applications. Gasification with a combination of steam and oxygen instead, can increase both the heating value of the producer gas (10–18 MJ/Nm^3) and the H_2/CO ratio. A high H_2/CO ratio is required for producing liquid fuels through synthesis.

However, the high costs for both oxygen supply equipment and operation are significant challenges for the commercial implementation of this technology.

Oxygen Transport Membranes:

Oxygen transport membranes (OTMs), the core technology investigated and developed in this project, have the potential to solve this challenges, as they are a promising alternative to produce oxygen for high temperature processes (like gasification) at much lower costs than via conventional routes (cryogenic distillation), especially for small to medium scale plants.

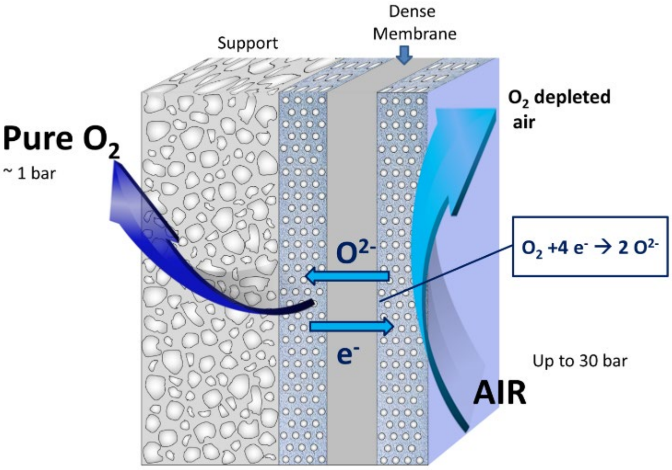


Figure 3.1 *Schematic illustration of on oxygen transport membrane*

In simple terms, an oxygen transport membrane is a gas-tight layer permeable only to oxygen, which allows “filtering” the oxygen out of air with a selectivity of 100 %. The key feature of an oxygen transport membrane is a gas-tight layer of a ceramic material, which can conduct both electrons and oxide ions, O^{2-}. On each side of the membrane, catalyst material is present in the activation layer to facilitate the splitting and recombination of gas molecules (Fig. 3.1) ($O2 + 4e^- \leftrightarrow 2O^{2-}$). The difference in oxygen concentration (partial pressure of oxygen (pO_2)) between the two sides of the membrane provides the driving force for the process. Hence, to deliver an oxygen flux an oxygen partial pressure gradient over the membrane must be established. This can be achieved by increasing the absolute pressure on the air side, by applying a vacuum on the low pO_2 side or purging here with a gas lean in oxygen (e.g. steam, CO_2) or even syngas. To maximize the oxygen flux through the membrane, advanced oxygen transport membranes typically have the architecture shown in Figure 3.1. They consist of a thin membrane layer (2 - 40 µm) placed on a thicker, porous structure providing the necessary mechanical support. The overall design may be planar or tubular, but for the use in biomass gasifiers tubular components, which give some benefits in terms of mechanical stability and are easier to integrate, are preferred.

To summarize: The most advanced and effective gasification schemes involve oxygen firing. Producing the needed oxygen in a cheap and efficient manner at an intermediate scale (plants of 25 kW_{th} - 100 MW_{th}) is crucial here, and one of the biggest hurdles is to make this concept commercially more attractive. High temperature ceramic oxygen transport membranes (OTMs) are a very promising technology to deliver this. Especially in small to medium scale process involving high temperatures, like the one addressed here, OTM technology is potently superior to other air separation techniques. To develop OTMs suitable for biomass gasification and to demonstrate their integration into a gasifier was the ambition of this project.

4. Project implementation

Project evolvement according to milestones:

Overall, the project evolved very well and all objectives and milestones (MSs) were reached. 14 out of the 17 milestones were reached within the originally anticipated timeframe, while the remaining three were full-filled with ~ six-month delay. The originally project duration of 36 month was extended by 11 month by two no-cost extensions. The first extension of six month was granted in May 2019, the reason being maternity leave of key personal; the second extension due to the COVID-19 lockdown of experimental facilities of five month was approved in April 2020.

An overview of all milestones, including status, date and eventual delays can found in Table 4.1, the latest version of the Gantt diagram can be found in the appendix.

Table 4.1 Overview of milestones in the *HighFlex* project

MS #	Month	Description	Status
MS 1.1.1	12	*Dip coating slurries and sintering program for all functional layers developed*	**Reached** as planned
MS 1.1.2	18	*Optimal component geometry selected for two stage gasifier, and membrane supports of that geometry extruded and characterized*	**Reached** as planned
MS 1.2.1	1	*Ceramic materials supplied to DTU by CoorsTek*	**Reached** as planned
MS 1.2.2	15	*Analysis of membrane fabrication at DTU performed*	**Reached** as planned
MS 1.2.3	32	*Membranes with a total of 4000 cm2 of active membrane area with reproducible properties fabricated*	**Reached** as planned
MS 2.1.1	12	*Rig for long-term-testing constructed and commissioned after internal safety approval*	**Reached**, 4 month delay
MS 2.1.2	24	*Membrane tested for 5000 h according to defined testing protocol*	**Reached** as planned
MS 2.1.3	32	*Membrane (from identical production batch used in MS 2.1.2) tested for 8500 h*	**Reached** as planned
MS 2.1.4	27	*Post mortem analysis on membrane tested in MS 2.1.2 performed and compared with results from non-tested sister membranes*	**Reached** as planned
MS 2.2.1	12	*Testing protocol for mechanical testing based on input from all partners established*	**Reached** as planned
MS 2.2.1	30	*Ceramic membranes developed in WP1 tested according to the protool defined in MS 2.3.1*	**Reached** as planned
MS 3.1.1	20	*Membrane performance for partial oxidation of tars determined*	**Reached** as planned
MS 3.1.2	22	*Best position for membrane integration in the TwoStage gasifier determined*	**Reached** as planned
MS 3.2.1	32	*Successful integration of a membrane tube module and proof-of-concept run with oxy-gen blown biomass gasification with the TwoStage gasifier at DTU*	**Reached**, 6 month delay
MS 3.2.2	32	*Recommendations and guidelines for a >100kW oxygen blown biomass gasifier design*	**Reached** as planned
MS 4.1.1	25	*Calculation of economic figures of merit for membrane integration in different integration schemes*	**Reached** as planned
MS 4.2.1	30	*Danish road map on optimal integration of oxygen blown biomass gasification including economic evaluation criteria and influence on the grid stability provided to public*	**Reached**, 6 month delay

Associated risks, experienced problems and unforeseen developments:

From the 11 possible risks identified before project, two were faced during the project. These were related to i) the membrane manufacturing (gas tightness), and ii) the problem to integrate OTMs into the biomass gasifiers. Both challenges could easily be solved by the suggested risks mitigation strategies. In summary, a few non-critical problems were encountered, but despite a temporary delay on some milestones, the *Highflex* project was not impacted by these encountered risks.

A small shift in the underling gasification technology in which the OTMs were integrated was suggested in M24 and implemented for the final measuring campaign. Instead of the suggested TwoStage gasifier, a new updraft pyrolysis unit was used to produce the raw gas. This pyrolysis unit is equivalent to the first stage (pyrolysis stage) of the TwoStage gasifier, thus the membranes were tested under comparable conditions. In retrospect, this decision was correct, as the updraft pyrolysis unit seems to be the more forward-pointing technology currently being scaled up.

The work task 4.2 “Integration of oxygen blown biomass gasification into the Danish energy grid” positively benefited from activities in other Danish national projects, mainly the ”Future Gas Project”, (funded ~ eight month later by Innovation Fund Denmark, https://futuregas.dk). The synergy between *HighFlex* and work package 4 in the Future Gas project (“WP 4 Gas in the integrated energy system”), and the fact that DTU Chemical Engineering and Evida participated in both project, allowed finishing task 4.2 with less resources (in terms of man hours) than originally anticipated.

5. Project results

The project was organized in four main activities: material development and up-scaling of oxygen transport membranes (OTMs) (WP1), long-term testing of OTMs (WP2), incorporation of OTMs for tar reduction in biomass gasifiers (WP3), and possible integration schemes for oxygen-blown biomass gasification in a future Danish energy system (WP4). In this report, technical result are reported along this WP structure. For each WP a short summary in layman’s terms is given, followed by more detailed summary of the main technical results relevant to the milestones and project objectives outlined above.

WP1 Membrane development

Summary: The overall aim of this work package was to establish a robust manufacturing route for optimized, high performance OTMs suitable for operation in biomass gasifiers. Industry-standard methods, such as extrusion and dip-coating, were used to manufacture the tubes. In a first step, the support geometry was optimized, and mechanical stable tubes were extruded. Afterwards, the functional layers were applied by dip-coating. Optimized materials with high performance and chemical stability were successfully implemented into the functional layers. After several process iterations and up-scaling steps, a reliable and industry-feasible fabrication protocol was established, which allowed to fabricate tubes with a length of >100 cm. The established process was analyzed with an industrial project partner, and was evaluated as sensible for up-scaled fabrication of tubular, asymmetric oxygen transport membranes.

The final, optimized architecture of the OTMs successfully produced in the *HighFlex project* is shown in Figure 5.1. The membranes had a tubular shape with a diameter of 1 cm and a length of up to 100 cm (Fig. 5.1, left). An overview cross section of such a tube with the different functional layers and materials used is shown in Figure 5.1 (right to middle). The single production steps and optimization iterations leading to this final product are outlined below.

Membrane fabrication:

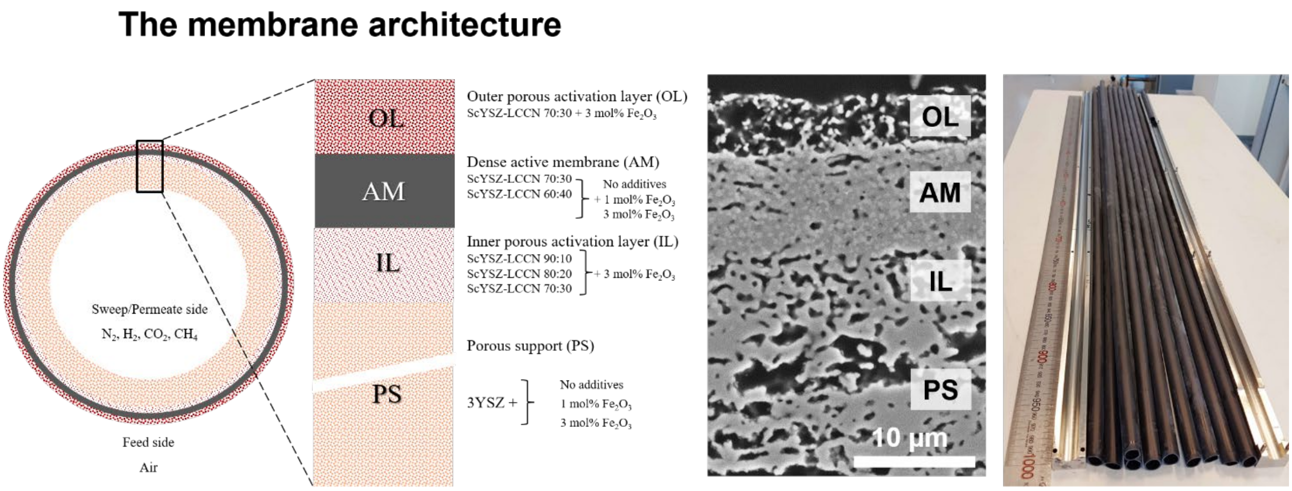


Figure 5.1 Illustration of the different functional layers within an OTM (left), SEM image of the optimized functional layers (middle), produced tubular OTM supports with 100 cm length (right)

The major production steps are outlined in Figure 5.2 below. Usually, three dip coating and co-sintering steps are repeated in between the extrusion and the infiltration of catalyst.

Figure 5.2. Major steps in the fabrication of tubular asymmetric OTM at DTU Energy.

As a first step, robust, tubular supports were produced via thermoplastic extrusion. $Y_{0.03}Zr_{0.97}O_{1.985}$ (3YSZ) was selected as the support material due to its high mechanical strength. The geometry of the extruded tubes was optimized for straightness, mechanical robustness and applicability in the selected gasifiers (M1.1.2). Straight tubes with 10 mm outer diameter after sintering and a wall thickness of 0.3 mm were found to be the optimum for the porous support. These tubes show, excellent mechanical strength (≥50 MPa) and high gas permeability (> $1x10^{-14}$ m^{-2}) after sintering. The used ceramic raw materials were standard industrial grade and were supplied by project partners (M1.2.1).

As second step in the manufacturing process, the functional layers (Fig 5.1) were applied by dip-coating onto the porous support. Here novel and more resistant membrane materials were evaluated for the targeted operation in biomass gasifiers (up to 1000 ºC, low pO_2 of down to 10^{-21} atm, presence of tar). Dual phase membranes based on $LaCr_{0.85}Cu_{0.1}Ni_{0.05}O_3$ (LCCN) and $(Sc_2O_3)_{0.10}(Y_2O_3)_{0.01}(ZrO_2)_{0.89}$ (ScYSZ) were found to have a good performance under these harsh condition.

Initially, functional layers, including the ScYSZ-LCCN based membrane, were deposited on the 3YSZ porous supports. Several severe challenges were faced during the manufacturing of these tubular membranes, such as extensively high processing temperatures (>1450 ºC) required to densify the membrane layer, accompanied by Cr vaporization and the formation secondary phases, which lead to fractures and cracks within the membranes.

By understanding the fundamental underlying processes and careful optimization of critical parameters in iterative loops, it was possible to overcome these challenges during the project. Successful modifications of the membrane architecture by using sintering aids (1 mol% and 3 mol% Fe_2O_3 to the porous support and the thin film membrane layer, respectively) reduced the stresses in the multilayer membrane caused by the sintering mismatches. The total shrinkage of the porous support was found to be the main driver for the densification of the membrane. Applying this new insight to the production processes (M1.1.1) allowed to decrease the sintering temperature by 200 °C and obtain a dense, crack free membrane layer after sintering at 1250 ºC, avoiding evaporation of Cr. Furthermore, the addition of Fe_2O_3 to the 3YSZ supports increased the strengths by a factor of 2.5 (>138 MPa) while still retaining an excellent gas permeability of $2.2x10^{-14}$ m^{-2}.

Once all manufacturing parameters were optimized, the up-scaling of the production of OTMs was carried successfully (MS 1.2.3), and OTMs were made available for testing in other WP2 and WP3. As shown in Figure 5.1, OTMs can now be manufactured on a routinely base in significant numbers. Supports with a length of 1 m can be extruded, and a reproducible procedure is in place for dip-coating and sintering. The target of producing membranes with an area of 4 000 cm^2 stated in MS 1.2.3 was easily reached, even exceeded as membranes with a combined active area of > 10 000 cm^2 were extruded during the project.

An analysis of the oxygen membrane fabrication at DTU Energy was performed in collaboration with CoorsTek Membrane Science (MS 1.2.2). The company visited DTU Energy at Risø campus and inspected the laboratories and fabrication facilities. A PhD student from DTU Energy performed his international research stay (3 month) as an internship at the company's facilities. The compiled report contains analyses of each fabrication step (thermoplastic extrusion, dip coating, co-sintering, infiltration of catalyst) and provides overall comments and factors to look out for, as well as suggestions for improvement of a few steps. Overall, the fabrication method employed by DTU Energy was found suitable for up-scaling to industrial fabrication volumes. The main difference between DTU Energy's process and common practice is the use of thermoplastic feedstocks. CoorsTek also pointed out the importance of monitoring the success rate of each process step, since this affects the total process yield. Most importantly, the feasibility of automating each step should be considered already at this stage, here the dip coating step (Fig. 5.2) was identified as most critical point in the DTU Energy procedure.

WP2 Demonstration of long-term stability and mechanical robustness

Summary: The aim of WP2 was to demonstrate that the membranes developed in WP1 have a sufficient life-time and mechanical robustness/reliability for real-life operations, both factors which are crucial to create confidence in the technology and create trust among its potential user. As a first step, a dedicated test rig was constructed ensuring sufficient test capacity for the long-term tests. Two types of OTMs were successfully tested for ~ 8 000 h. OTMs based on ScYSZ showed an excellent stability, and no degradation neither in performance nor in the microstructure was found. Additionally, the mechanical stability of the OTMs was measured, and excellent mechanical strength, surpassing the target by a factor of >2, was demonstrated.

A special rig for long-term testing in different gases (CO_2, N_2, H_2/N_2) was designed, constructed and commissioned (MS 2.1.1) (Figure 5.3). It consists of three separate furnace chambers, such that three membranes can be run in parallel, and exchanged independently, e.g. in case one of tested membranes fails. Safety-related authorizations took longer than planned and let to a 4 month delay of MS 2.1.1. The test conditions, such as temperature, gas composition, flow rates and heating profiles, were defined together with the (industrial) partners.

Long term stability tests of selected membranes based on $CaTi_{0.85}Fe_{0.15}O_3$ (CTF) and ScYSZ based membranes (see WP1) were tested for approximately 8 000 hours (MS 2.1.2, MS 2.1.3) using N_2/air and CO_2/air gradients. The performance of the membranes is shown in Figure 5.4. The CTF tube (red line) show a variation of approximately -25% of the initial O_2 flux, but performed very stable without a significant degradation. OTMs based on ScYSZ (green line) started at a lower oxygen production value initially, but reached a similar performance as the CTF tubes after 1 000 h. After that point, also this membrane performed very well. In conclusion, both membranes did not show any signs of degradation after almost 8 000 h of operation in CO_2 or N_2, which is a very promising result.

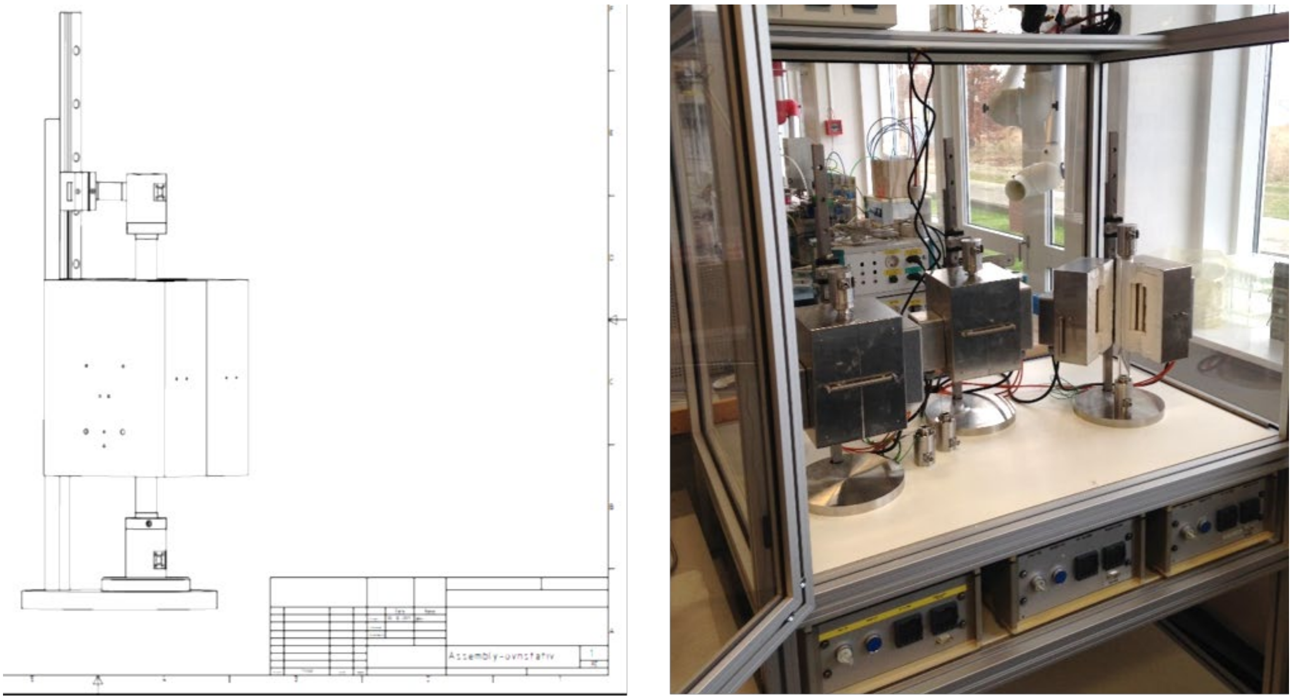


Figure 5.3. Left: Sketch of one of the three furnaces for long-term testing of OTM tubes. Right: Picture of the constructed long-term testing rig with three furnaces (one opened on the right hand side) for simultaneous testing of three individual OTM tubes.

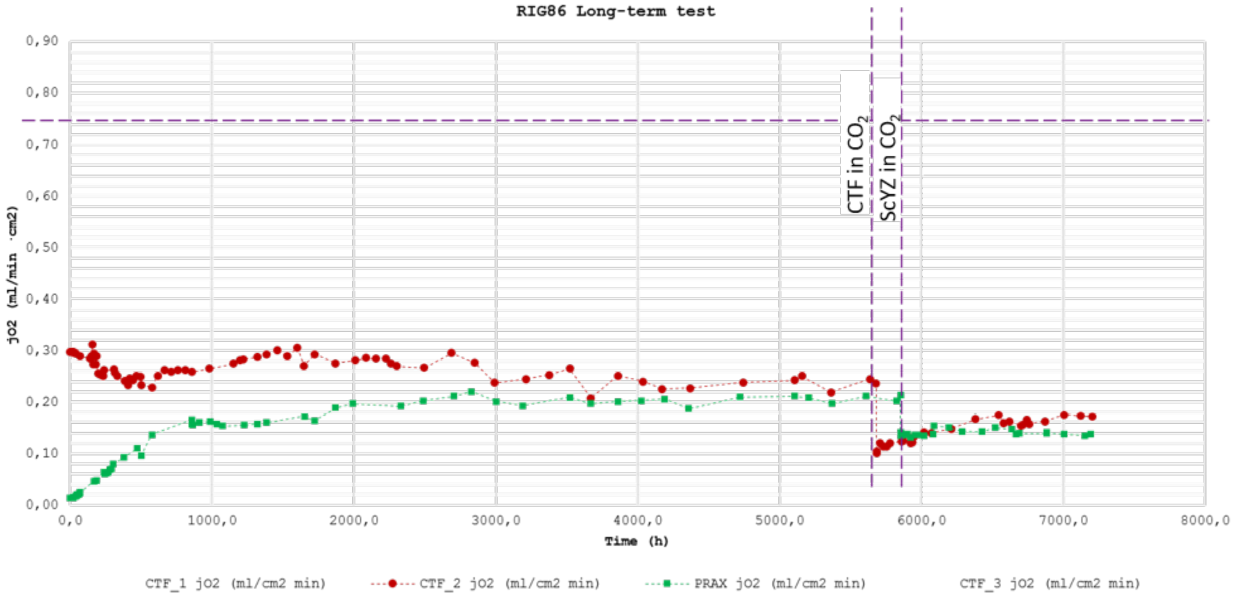


Figure 5.4 Long-term testing of CTF (red) and ScYSZ (green) based OTMs. The oxygen production is plotted over time. No degradation was observed.

The microstructures of the tubular membranes tested for approximately 8 000 h were analyzed post-mortem (MS 2.1.4) on fractured and polished cross-sections by scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX), and compared to untested membranes. No significant microstructural degradations were found, which is expected based on the stable performance reported above.

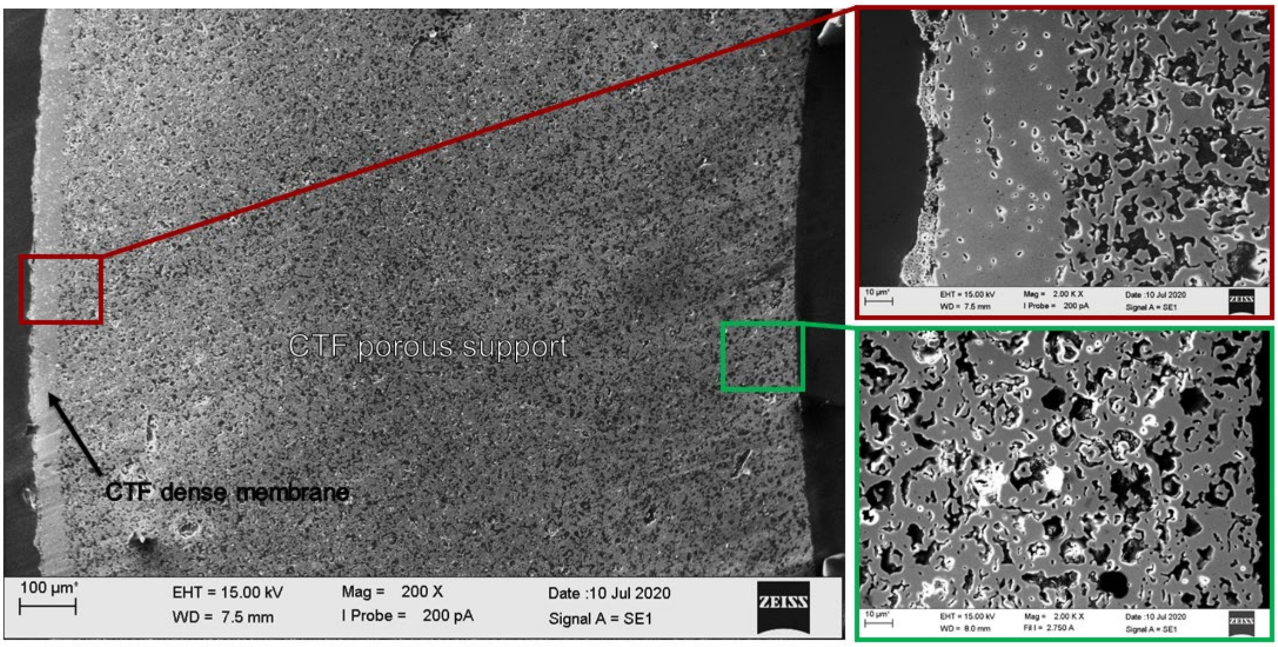
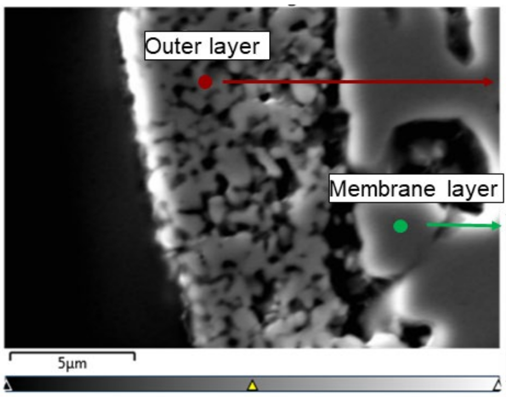


Figure 5.5 SEM images of post-mortem CTF tubular membranes tested over 7 250 h.



Element	Wt%	Wt% Sigma
C	8,14	0,71
O	31,78	1,21
Si	0,97	0,15
Ca	26,51	0,67
Ti	29,72	0,79
Fe	2,87	0,49
Total:	100	

Element	Wt%	Wt% Sigma
C	6,02	0,6
O	29,2	1,21
Ca	29,03	0,7
Ti	31,99	0,8
Fe	3,77	0,48
Total:	100	

***Figure 5.6** Schematic illustration of on oxygen transport membrane2.3: EDX analyses performed on the outer and membrane layers of CTF tubular membranes.*

SEM micrographs of the post-mortem CTF asymmetric membranes are presented in Figure 5.5. The membranes do not suffer from any significant contamination or degradation. However, EDX analyses presented in Figure 5.6 shows that trace of Si can be found in the outer porous layer of the asymmetric membranes, but not on the active dense membrane layer.

Similarly, the SEM analyses did not reveal any degradation or contamination of the tested ScYZ based membranes. SEM

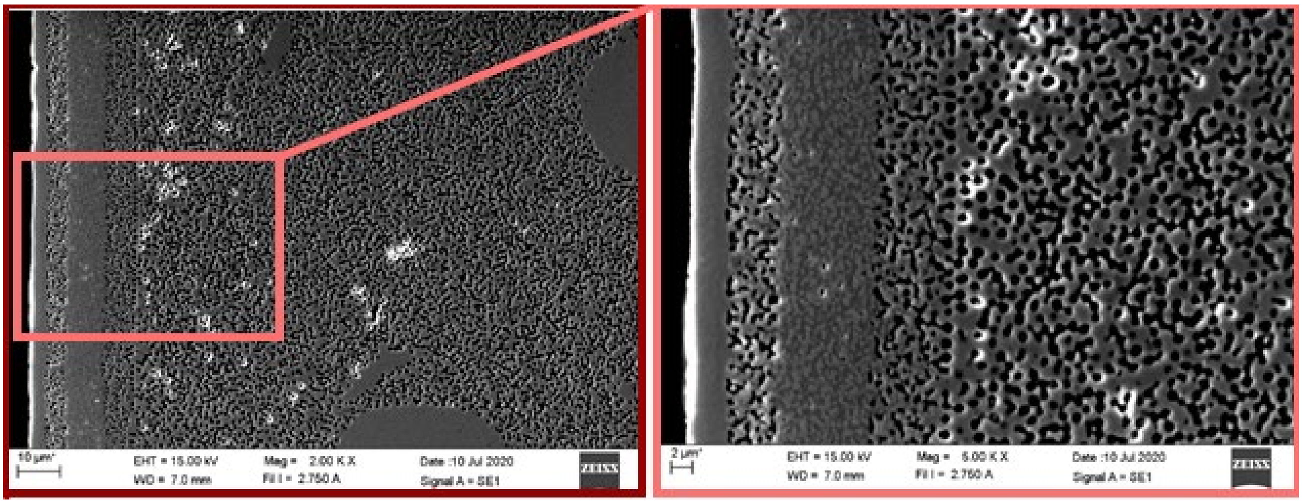


Figure 5.7 Post-mortem SEM images of ScYZ based tubular membrane

images presented in Figure 5.7 show a global view of the tubular membrane and closer views of the dense membrane layer and of the porous support. No changes or damages in the microstructure were observed after testing, and no contamination were detected by the EDX system.

It should be pointed out that comparable data on long-term testing on asymmetric oxygen membrane for such long time (8 000) h have not been reported before. Indeed, having the capability to test membranes for such long times strengthened DTU Energy's attractiveness as partner in future projects, for scientific consortia as well as for industry as hoped for, and already during the project period new industrial partners (Linde/Praxair, US) were attracted, underlying the importance of the test rig also for future projects.

Next to the long-term stability of the oxygen production, also the mechanical robustness of the produced OTMs was investigated. A mechanical testing protocol was developed for testing of tubular species at DTU Energy (MS2.2.1). It covers flexural strength measurement using four-point bending setup with two support pins separated by 50.0 mm and two loading pins separated by 25.0 mm (Fig. 5.8). The tubular specimens are to be cut to a length of 60 mm with straight end surfaces. Diameter and thickness were measured with 0.01 mm accuracy for calculation of strength and effective volumes. The tubes are then cut along their length into two sections covering 180° sectors of the tube.

The two inner pins in the four-point bending setup (Fig. 5.8, left) presses the cut edges, which together with the specimen geometry will ensure the fracture origins from the zone of tensile stress in the middle of the specimen, unaffected by stress from cutting defects. The strength of up to 30 samples from each relevant type of OTM support was measured for good statistical analysis. This high number of tests was made possible with a unique in-house constructed loading rig where 16 samples are simultaneously mounted (Fig 5.8, right). In this manner, the required 30 specimens could be tested with only two test sequences.

A minimum flexural strength of 50 MPa was defined as the target for the tubular supports to be used in target application. This strength requirements is displayed as dotted red line in Figure 5.9.

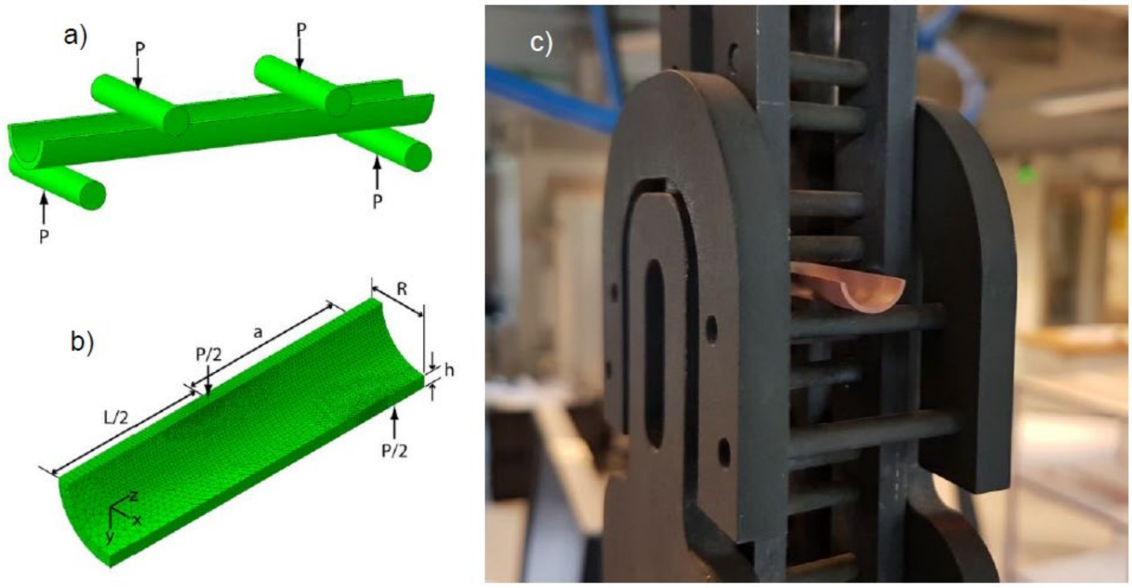


Fig. 4.14. 4-point bending testing. a) Test configuration of 4-point bending of a semi-cylindrical specimen, 2) finate element mesh of a quarter of specimen, c) mounting of a 3YSZ sample in the testing rig. [40]

Figure 5.8 Test configuration of 4-point bending of a semi-cylindrical specimen (left), mounting of a 3YSZ sample in the testing rig (right).

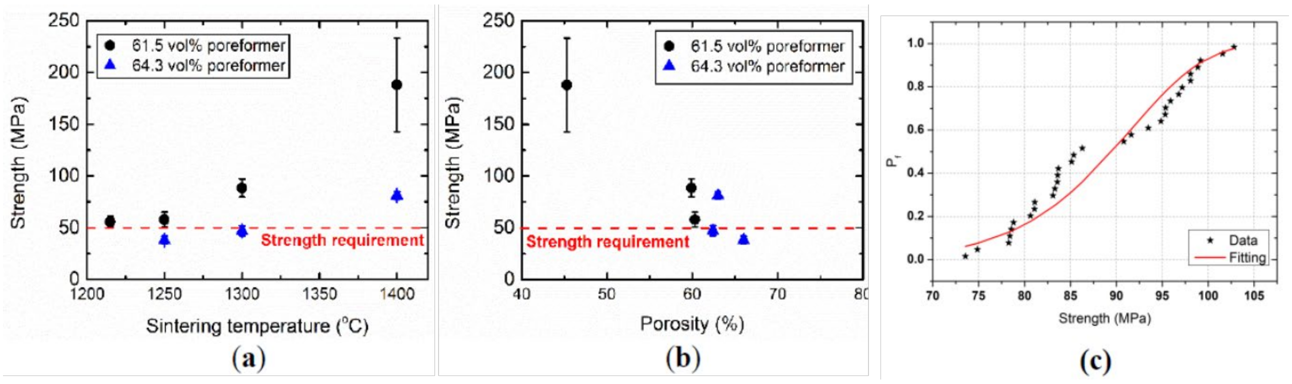


Figure 5.9 Strength measurements of OTM supports as function of sintering temperature (a), and porosity (b), and the Weibull plot for room temperature strength of OTM supports (c)

Figure 5.9 shows strength vs. sintering temperature and porosity of the two relevant porous 3YSZ supports manufactured in this project (MS 2.2.2). Both compositions show a porosity content which is in good agreement with the amount of pore former, and decreasing strength with increasing porosity, as expected. While the tubes made with 64.3 vol% pore formers are too fragile at low sintering temperatures, the ones with 61.5 vol% pore formers fulfil the desired strength of 50 MPa after sintering in the entire studied sintering temperature range. The 61.5 vol% pore former composition was therefore chosen for further development of the asymmetric OTM in this project (see WP1). Figure 5.9 (c) shows the Weibull plot for room temperature strength of the specimens with 61.5 vol% pore former and sintered at 1300 °C. The measured strengths of the specimens are shown with the assigned probabilities of failure (P_f) and a fit of a Weibull distribution. Results of the Weibull fit show that the specimens have a characteristic strength and Weibull modulus of 92.19 MPa and 12.20, respectively. Further increase in the flexural strength could be achieved by using small amount of Fe_2O_3 as sintering aid. By using 1 mol% Fe_2O_3, the flexural strength of the 3YSZ support increased by 38% up to 112 MPa, and even higher values of >138 MPa were calculated for 3YSZ support containing 3 mol% Fe_2O_3, which is almost times higher than the initial defined project target of 50 MPa

WP3 Integration and demonstration of OTMs in biomass gasification processes

Summary: The goal of WP3 was to demonstrate the capability of the membranes developed in WP1 to operate in real biomass gasification facilities. First-of-its kind tests in the gasifier were carried out in this WP, in which OTMs where used to eliminate tars form the producer gas. Analysis of the gas and the tar composition at the outlet of the membrane demonstrated that a) it contributed to the partial oxidation of the tars, and b) that the fraction of useful gases could be increased.

Two test campaigns were run in the *HighFlex* project in November 2017 and December 2020 to demonstrate OTM operation the in real biomass gasification facilities.

A specific open flow setup (Fig 5.10) was designed to carry out tests with H_2, N_2 or producer gas from a side stream of the LT-CFB gasifier. The membrane was placed in the setup with the inner side as the permeate side and the outer side as a feed side in the hot zone of the furnace, which was heated up to 850˚C. The active membrane area was ca. 35 cm^2.

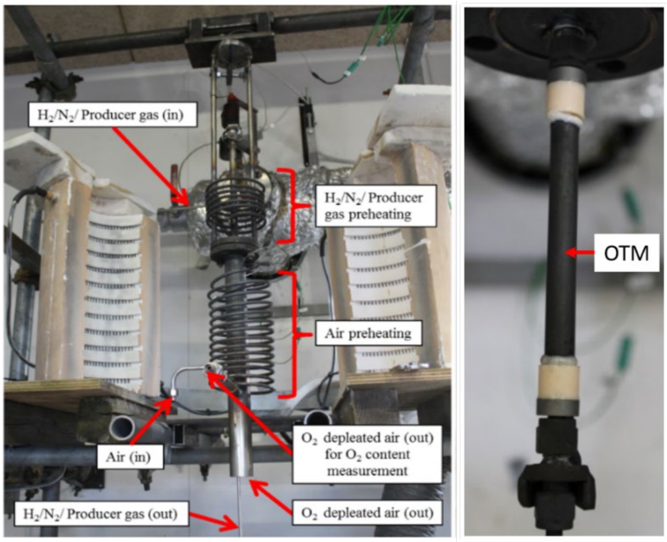


Figure 5.10 Set-up for the oxygen membrane testing on a side stream of the LT-CFB gasifier (left), oxygen membrane placed in the set-up unit (right)

In the first campaign, $Ce_{0.9}Gd_{0.1}O_{1.95}$ – $La_{0.6}Sr_{0.4}FeO_3$ (CGO-LSF) membranes, which were available at the project start, were tested for the partial reduction of tars. Crushed wheat straw pellets were used as fuel for the combustion in the LT-CFB gasifier. The OTM performance tests and tar decomposition measurements in the producer gas were started when stable operation conditions were reached ca. 4 hours after the start-up. Samples of the producer gas tars were taken at the input and output streams of the membrane unit for analysis. Additionally, gas samples for off-line analysis were taken directly at the output stream of the partial oxidation unit.

The membrane was first tested in N_2, then in producer gas for ca.120 min, and finally in N_2 again to determine the degradation of the membrane. The initial performance of the membrane in N_2 was an oxygen flux of 0.9 Nml·cm^{-2}·min^{-1}. After the test in producer gas, the flux in N_2 reduced to ca. 0.5 Nml·cm^{-2}·min^{-1} (Fig 5.11). Post-mortem analysis of the membranes revealed that the performance decrease after exposure to the producer gas is most likely related to the deposition of carbon and sulphur-based compounds in the porous support in the inlet area of the membrane tube.

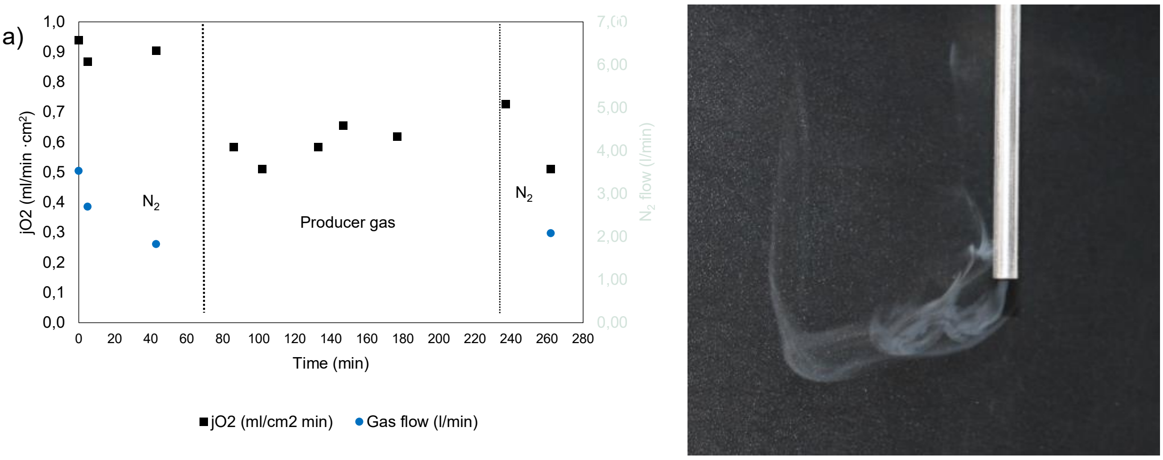


Fig. 5.11 a) Oxygen permeation flux of a 35 cm^2 CGO-LSF membrane tested in the partial oxidation test unit adjacent to a LT-CFB gasifier at 850 ˚C. b) White producer gas at the output of the membrane indicating the partial oxidation of the tars.

A detailed analysis of tar composition before and after the partial oxidation unit is shown in Figure 5.12. Phenols and naphthalenes are fully decomposed when the gas reached the outlet of the membrane, but tertiary tars (PAH), such as fluorene, anthracene and pyrene were still identified. This indicates that partial oxidation of tars took place. To determine if tar conversion is thermally driven or can be ascribed to the oxygen supplied by the OTM, a control experiment with a dense alumina tube, which does not provide oxygen to the producer gas, was carried out under similar conditions. Similar as when operating the OTM, mainly tertiary tars (PAH) were observed at the outlet, indicating that also thermal decomposition played a role.

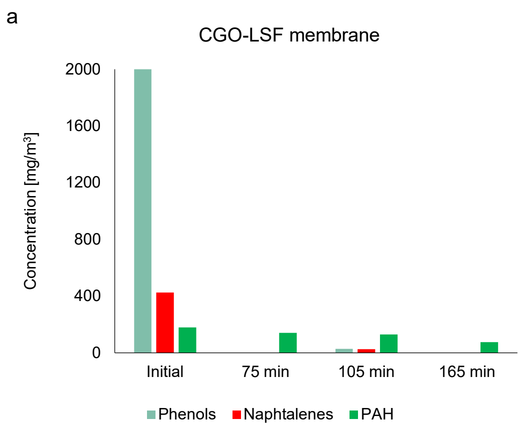


Figure 5.12 Tars composition after treatment in a partial oxidation unit using a CGO-LSF membrane.

However, it is noteworthy that the concentration of tertiary tars in the OTM experiments is lower than that obtained in the control experiment with the alumina tube, even if the initial concentration of total phenols was higher in the OTM test. When comparing the oxygen flux and formation of tertiary tars, a correlation between the values is observed, which indicates that the tars conversion is partly linked to the oxygen introduced via the OTM (MS 3.1.1).

Next to the decomposition of tars, also the gas composition before and after passing through the OTM was analyzed. Again, similar experiments were performed with an alumina tube as control. It is found that gases such as H_2, CH_4 and CO are generated in both experiment (OTM and control), which confirms that also the thermal processes contribute to the partial decomposition of tars. Nevertheless, in the OTM experiment, ca. 50% and 20% more H_2 and CH_4 respectively are obtained compared to the control group, while the concentration of CO remains similar. Also the relative concentrations of N_2 (and CO_2) decreases stronger for the membrane case, even after correcting the values for the effects of the unintended difference in inlet tar content.

In the second campaign (December 2020), several improvements were implemented. A new updraft pyrolysis unit with a new full-stream partial oxidation reactor (POX-reactor) unit was used. The OTM membranes were directly integrated in the POX-reactor for tar reforming of the pyrolysis gas (MS 3.2.1). The membrane module was built from commercial available parts, the used membranes in the reactor were ScYZ based (see WP1), and where much more robust than the ones used in the 1st campaign. Also the membrane area was scaled up by 600 % from 35 cm^2 to 220 cm^2. Again a control group experiment was run in parallel. The gas composition at the membrane outlet (Fig 5.13) was stable throughout the experiment, and no decrease in the performance of the membrane before and after the experiments was detected (compared to >50 % decrease in 1st campaign). The analysis of the tars in the producer gas before and after passing through the membrane showed that more than 98% of the tars were removed.

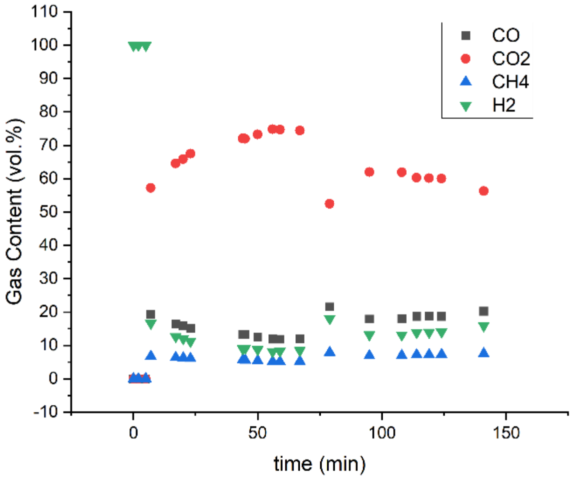


Figure 5.13 Fraction of relvant gases in the producer gas after OTM

In conclusion, data from the measurements campaigns showed that the oxygen permeating through the OTM into the producer gas contributes to the tar decomposition via a partial oxidation, and helps to increase the gas quality significantly. Using improved, more robust membranes could significantly enhance the stability and the amount of tar reduction. No performance degradation of the membrane during the tests was detected, and >98 % of tars were removed – all in all very promising results exceeding initial expectations.

In parallel, the integration of oxygen transport membranes in the two gasification concepts developed at DTU and their possible scale up options for oxygen blown operation have been theoretically studied (MS 3.2.1 and MS 3.2.2). The two gasification concepts developed at the Biomass Gasification Group of the DTU Chemical Engineering Department are a TwoStage gasification process and a Low Temperature Circulating Fluidized Bed gasifier (LT-CFB). Both have been tested in air-blown mode in several experimental campaigns with different biomasses. Recently, they have also been successfully tested with O_2/CO_2 mixtures as gasifying agent.

The tests and experimental campaigns ran in the *HighFlex* project showed that it is possible to direct integrate OTMs with biomass gasification processes, but the integration is highly dependent on the performance and stability of the membranes when exposed to the harsh environment inside the gasifiers. In summary, the recent conducted studies suggest that the best placing for direct integration of OTMs and scaling up the systems are:

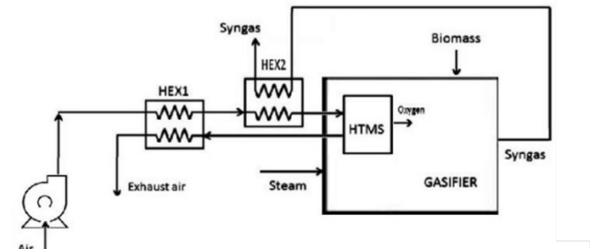
For the TwoStage gasification process: This type of reactor should be scaled up as two separated updraft reactors and the oxygen membranes should be placed in the partial oxidation zone after the first reactor (MS 3.1.2). For the LT-CFB gasifier system, which has already been scaled up to 6MW, the integration scheme for oxygen membranes is different. Here the best placement for OTMs would be in an external module for oxygen production or in a POX reactor for tar reforming and cleaning. In both cases, larger proof-of-concept modules and longer operation time in real gasifiers are recommended as the next logical step to test the operation of OTMs membranes in even more realistic conditions.

WP 4: Techno-economic assessment of oxygen blown biomass gasification

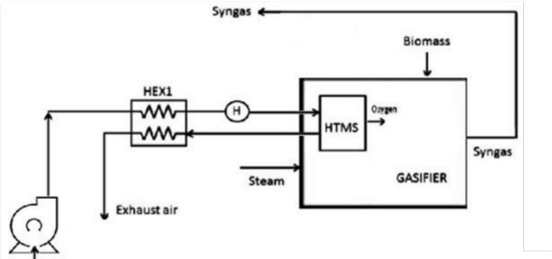
Summary: The outcome of WP4 is an initial techno-economic analysis of the integration of OTMs in different biomass gasifier designs based on the outcome of WP1and WP3. It was found that in comparison to state-of-the-art cryogenic plants for oxygen production, OTMs seem to be a promising and cost competitive alternative. Additionally, the public report "Danish roadmap for sustainable gas grid transition – status of the role of thermal gasification" was published. In this report the current situation and future scenario potential of especially SNG production from biomass gasification was analyzed, based on recent publications and more novel input from different stakeholders in the project consortium.

Techno-economic analysis of integration of membranes in different biomass gasifier designs:

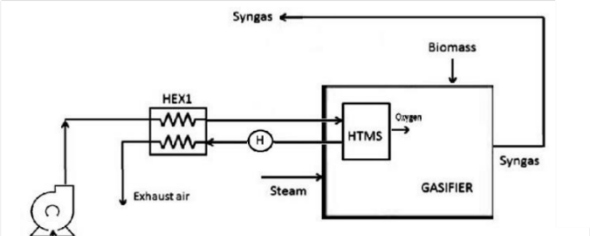
Based on the results from WP3, three different configurations were investigated for direct integration of oxygen transport membranes (OTMs) inside a biomass gasifier to provide O_2 for the gasification process (Fig.5.15).



Configuration Direct Bio1



Configuration Direct Bio 2



Configuration Direct Bio 3

Fig. 5.15 Configurations studied for direct integration of the membranes inside the biomass gasifier

As a first step, the energy consumption related to the oxygen producing of these new configurations (“Direct Bio 1-3”) was calculated, and compared with configurations from a previous study, which did not consider the direct integration of the OTMs. It was found that configuration “Bio Direct 1” (Fig. 5.16, left, black line) can achieve significant lower energy consumption values than previous analyzed configurations (~ 200 kWh/tO_2), both at 750 and 850˚C. The energy consumptions of the configurations “Bio Direct 2” (Fig. 5.16, middle, black line) and 3 (Fig. 5.16, right, black line) were found to be in the range of the “best cases” achieved in previous studies.

Next to the energy consumptions, also the membrane area required was calculated. This area strongly depends on the kind of membrane selected. In this study, two types of membranes were considered: OTMs based on ScYZ-LCCN developed in WP1, and “ideal” OTMs based on $Ce_{0.9}Gd_{0.1}O_{1.95}$ (CGO), which represent a “best case scenario” theoretical possible. Results are included in Figure 5.16 as blue lines, and were also used for estimating the total plant investment costs.

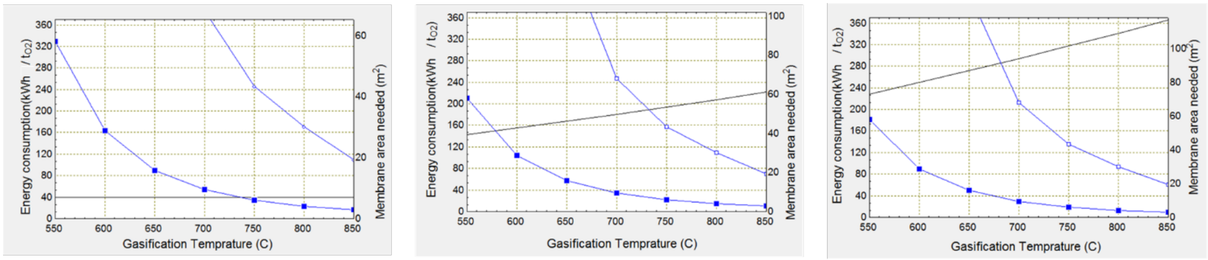


Figure 5.16 Energy consumption (black line) and membrane area (blue lines) needed for a 6 MW_{th} biomass gasification plant with Configuration Direct Bio 1 (left), Configuration Direct Bio 2 (middle), and Configuration Direct Bio 3 (right) at different gasification temperatures. Air feed pressure is equal to 1 bar. Filled markers correspond to CGO and hollow ones to ScYZ-based OTMs.

In addition to the energy demand and membrane area required, the total plant investment (TPI) and variable operating and maintenance (VOM) costs have also been estimated (MS 4.1.1) and are summarized in Table 5.1. The results obtained for TPI and VOM show lower costs for any of the three configurations compared to the reference case of a cryogenic distillation plant. Configuration Direct Bio 1 has the highest TPI costs because it has two Inconel heat exchangers. The TPI cost differences between Configuration Direct Bio 2 and 3 are due to Conf. 2 having a furnace before the membrane and Conf. 3 having just a burner after the membrane. Regarding the variable operating and maintenance costs, Configuration Direct Bio 1 has the lowest costs because it does not have any burner/furnace that consumes natural gas or any kind of fuel.

Table 5.1. Total plant investment (TPI) and annual variable and operating maintenance cost (VOM) for the different configurations studied, using a cryogenic distillation plant as a reference case.

Total plant investment (TPI)				
(k$)	**Conf. Direct Bio 1**	**Conf. Direct Bio 2**	**Conf. Direct Bio 3**	**Cryogenic plant**
Equipment costs (EC)	799.8	610.2	559.2	1505.8
Instrumentation & electromechanical control (IEC)	80.0	61.0	55.9	
Fixed costs (FC)	879.8	671.2	615.1	
Base plant costs (BPC)	985.3	751.7	688.9	
Project Consistency (PC)	197.1	150.3	137.8	

Total facilities investments (TFI)	1182.4	902.1	826.7	
Startup cost(SC)	20.9	58.4	83.2	
TPI	**1203.3**	**960.4**	**909.9**	**1505.8**
Annual variable operating and maintenance costs (VOM)				
Contract and material maintenance costs (CMC)	59.1	45.1	41.3	1911.3
Local taxes and insurance (LTI)	17.7	13.5	12.4	
Labor costs (LC)- 1 person/shift 4 shifts (8 h/day) 17$/h	127.5	127.5	127.5	
Membrane replacement costs (MRC)	0.7	0.7	0.7	
Utility costs (UC)	3.8	396.7	650.0	
VOM	**208.9**	**583.6**	**832.0**	**1911.3**

In conclusion: Even though a complete and more detailed economic analysis would be required, Configuration Bio Direct 1 seems to be the best option for direct integration of OTMS inside a biomass gasifier. The practical realization of this configuration would require that membranes can withstand the harsh conditions present in the gasifier, which seems possible based on results from WP3. In comparison to state-of-the-art cryogenic plants for oxygen production, OTMs seem to be a promising and cost competitive alternative.

The second task in WP4 was related to creating of a first version of a roadmap for the development and implementation of (oxygen blown) biomass gasifiers in the Danish energy/gas grid (MS 4.2.1). The final *"Danish roadmap for sustainable gas grid transition – status of the role of thermal gasification*" is publically available here and on the homepage of the partnerskab for Termisk Forgasning) built on existing work from previous projects on conventional biomass gasifiers, as well as on the assessments of the short term and longer term role of biomass gasification evaluated in this project.

The report aims at assessing the current situation and future scenario potential of especially the option of SNG production from biomass gasification, and summarizes the visions of future gas grid scenarios according to publications and novel input from the different stakeholders. The main findings can be summarized as followed: A decrease in gas consumption of 40% towards 2030 and 54% towards 2040 is expected, while the share of green gas in Danish gas system is expected to be around 63% in 2030 and 100% in 2040.

Methanation of syngas deriving from thermal biomass gasification is a viable alternative to the use of fossil fuels and it has the great advantage of producing SNG which could be directly injected into the distribution grid and sold. However, this process involves additional costs that, nowadays, make it more expensive than the cost of methane from biogas plants. However, since this is a less mature technology than anaerobic digestion, thermal gasification offers greater potential for technological innovation and cost reductions. The use of oxygen transport membranes in biomass gasification would reduce the costs of oxygen supply for oxygen-blown biomass gasification and as a result for SNG production. Two conceptual plant options for integration of SNG production in highly fuel-flexible, but still highly effective, central and decentral CHP plants have been suggested for further evaluations.

6. Utilization of project results

The *Highflex* project started as research project under the former Forskel PSO funding scheme, and consequently the main focus of the project was on R&D rather than on creating commercial results. The TRL of the core technology developed ("OTMs for biomass gasification") increased as planned from 3 to 5 during this project, but still further field test on a larger scale are considered relevant before market entry/commercialization. But despite the low TRL, several points to be addressed before commercialization have already been identified.

Competing solutions: Oxygen produced by (solid oxide) electrolysis

During electrolysis oxygen and hydrogen are produced simultaneously, and in principal both gases can be used to enhance the gasification process. The large increase in installed electrolysis units expected in the next years will lead to the availability of large amount of oxygen (as by-product), which could be a possible entry barrier for OTMs. A careful analysis how these technologies are competing or complementing each other needs to be carried out. The fundamental differences between the two technologies is are that

a) using the oxygen produced by electrolysis to produce sustainable fuels (e.g. methanol or methane) takes place when there is a surplus of electricity (e.g. from wind power), while
b) the high efficient conversion of biomass with OTM technology (the core of this project) to produce electricity takes place most economically when electricity demand exceeds production.

Therefore, the interaction of these two complementary technologies could be ideal to balance the (Danish) energy grid while ensuring the most efficient use of the limited biomass resources. However, if oxygen produced by electrolysis in time of electricity surplus could be stored in large amounts, which is not possible in an energy efficient manner today, and used when needed, electrolysis could become the preferred solution. Consequently, the lowest (market) entry point for OTMs is expected to be in areas with low electricity surplus/high electricity prices and large availability of biomass. Finland, Sweden or Canada would be examples for such locations.

Utilization of PhD project results in teaching and dissemination activities.

One PhD student was involved in the *HighFlex* project. His work resulted in manuscripts for six publications and seven conference contributions (listed in the appendix). Parts of the PhD work were used for teaching in two courses at DTU related to biomass and hydrogen production.

Utilization of technological results obtained project in the future:

The technical results obtained in *HighFlex* will be directly used in large EU project recently granted under H2020 Call: H2020-LC-SC3-2018-2019-2020, BUILDING A LOW-CARBON, CLIMATE RESILIENT FUTURE: SECURE, CLEAN AND EFFICIENT ENERGY. This EU project (Flexible Production of Synthetic Natural Gas and Biochar via Gasification of Biomass and Waste Feedstocks (FlexSNG)) is coordinated by VTT, Finland, and will start in 06/2021. DTU will lead the work package on "low cost oxygen production" and will be responsible for the "*Design and construction of a proof-of-concept oxygen transport membrane module*" and a "*Cost assessment and sensitivity analysis for a commercial-scale industrial OTM module*". These activities are based directly on results obtained in *HighFlex*, and the collaboration established with Linde/Praxair will continue in FlexSNG, ensuring a seamless continuation of the work. In summary, the next relevant steps for bringing OTM technology closer to market, industrial involvement, up-scaling, detailed cost assessments and field-test demonstration, will be addressed in this follow-up project.

The biomass pyrolysis technology applied in the last measurement campaign is currently scaled up to MW scale by the Danish company Stiesdal A/S (https://www.stiesdal.com/carbon-negative-fuels/); this scaling up is not related to the *HighFlex* project, but it should be noted that OTMs are among the possible alternatives discussed for oxygen delivery to boost the biomass pyrolysis process and syngas production in the future.

7. Project conclusion and perspective

Main results and conclusions made in the project: The main objective of the *HighFlex* was to demonstrate that oxygen transport membranes (OTMs) can be directly integrated into biomass gasifiers, and to show that such an integration increases the quality of the produced syngas in a cost-effective manner. This objective was successfully reached. A new generation of OTMs for the targeted application was developed, optimized materials with high performance and chemical stability were successfully implemented, and a reliable and industry-feasible fabrication protocol was established. The optimized manufacturing process for this new generation of membranes is now available for future follow–up project which will bring this technology closer to market. Furthermore, the developed OTMs showed sufficient life-time and mechanical robustness/reliability for real-life operations, both factors considered crucial to create confidence in the technology and create trust among its potential user. First-of-its kind tests demonstrating the capability of the membranes developed in *HighFlex* to operate in real biomass gasification facilities should be highlighted. Data from these measurement campaigns showed that the oxygen permeating through the OTMs into the producer gas contributes to the decomposition of undesired tars (>98 % of tars were removed), and helps to increase the gas quality significantly. The initial techno-economic analysis of the integration of OTMs in different biomass gasifier designs found that in comparison to state-of-the-art cryogenic plants for oxygen production, OTMs seem to be a promising and cost competitive alternative.

Next steps for the developed technology: The highly efficient use of biomass to generate electricity, heat, synthetic natural gas and transport fuel will be important for a future Danish energy system based on 100 % renewables. Two conceptual plant options for integration of SNG production in highly fuel-flexible, but still highly effective, central and decentral CHP plants have been suggested for further evaluations *in the public report "Danish roadmap for sustainable gas grid transition – status of the role of thermal gasification", which was compiled in this project. A direct continuation of the development efforts after the project end has been assured by DTU´s participation in a large follow-up EU project (FlexSNG), which will start in 2021.*

8. Appendices

Dissemination

The project has gained both national and international interest, demonstrated by invited talks both at world-leading ceramics conferences and in Ingeniørforeningen (IDA Mechanics) and awards for best presentations. Some of the results are already published, and several manuscripts are in their final preparation stage. All dissemination activities are listed in this Appendix.

A.B. Haugen (invited speaker) "Ceramic processing of tubular, multilayered oxygen transport membranes" 41st International Conference and Expo on Advanced Ceramics and Composites, Daytona Beach, US, January 2017

Andreas Kaiser (invited speaker), Astri Bjørnetun Haugen, Wenjing (Angela) Zhang, Simona Ovtar, Wolff-Ragnar Kiebach, Peter Vang Hendriksen. “Advanced manufacturing of porous ceramic structures for use in energy applications”, ECerS 2017 - 15th Conference & Exhibition of the European Ceramic Society, Hungary, June 2017.

A.B. Haugen (invited speaker). “Keramiske membraner til iltblæst forgasning”. Udvikling af fremtidige metoder til produktion af biobrændstoffer og grøn energi, IDA Mechanical, Aarhus, November 2017.

* Lev Martinez Aguilera (poster presenter), Astri Bjørnetun Haugen, Wolff-Ragnar Kiebach, ”Dual phase composites for tubular oxygen transport membranes”, Sustain conference, Denmark, December 2017. * Awarded prize for the best poster.

Martinez, L.; Haugen, A.B.; Kiebach, R. *Dual phase composites for oxygen transport membranes*. Poster presentation. DTU Energy's PhD Symposium. Denmark, 2017.

Martinez, L.; Haugen, A.B.; Kiebach, R. *Development of oxygen tubular oxygen transport membranes based on dual-phase chromites and ion-conductors*. Oral presentation. 2nd Nordic Conference on Ceramic and Glass Technology. Denmark, 2018.

Martinez, L.; Haugen, A.B.; Kiebach, R. *Compatibility Analysis of Chromites in Dual-Phase Composites for Oxygen Transport Membranes*. Poster presentation. The Electrochemical Society. Americas International Meeting on Electrochemistry and Solid State Science (AiMES). Mexico, 2018.

* Martinez, L.; Pirou, S.; Haugen, A.B.; Kiebach, R. *Stability analysis of chromite-based dual-phase composites for oxygen transport membranes*. Poster presentation. Fraunhofer, 15th International Conference on Inorganic Membranes. (ICIM). Germany, 2018 * Awarded prize for the best poster.

Astri Bjørnetun Haugen*, Lev Martinez Aguilera, Kawai Kwok, Tesfaye Molla, Kjeld Bøhm Andersen, Stéven Pirou, Andreas Kaiser, Peter Vang Hendriksen, Ragnar Kiebach, “Exploring the Processing of Tubular Chromite- and Zirconia-Based Oxygen Transport Membranes”, Ceramics, 1 [2], 2018

Puig-Arnavat, M.; L. Martinez Aguilera, L.; Otvar, S.; Sárossy, Z.; Bjørnetun Haugen, A.; Kiebachb, W.R.; Ahrenfeldt, J.; ‘Henriksen, U.B.; Hendriksen, P.V. (2018) Tar Reduction in Producer Gas by Use of Oxygen Transport Membranes. 26th European Biomass Conference and Exhibition. 14-18 May 2018 – Copenhagen, Denmark. (Oral presentation)

Ragnar Kiebach, Simona Ovtar, Stéven Pirou, Shiyang Chen, Astri Haugen, Jonas Gurauskis, Andreas Kaiser, Peter V. Hendriksen. Ceramic composite membranes for oxygen separation: a short review on recent developments and challenges. 15th international conference on inorganic membranes, 2018, Dresden, Germany.

Mohammadi, S. (2019) MIEC simulation for direct integration in gasification plant. Master Thesis. University of Bologna, Chemical and Process Engineering Department

Martinez, L.; Haugen, A.B.; Kiebach, R. *Tubular oxygen transport membranes based on dual phase composites*. Poster presentation. The 22nd International Conference on Solid State Ionics (SSI-22). Republic of Korea, 2019.

Martinez, L.; Haugen, A.B.; Kiebach, R. *Manufacturing of highly stable ScYSZ-LCCN tubular oxygen transport membranes*. Oral presentation. DTU Energy's PhD Symposium. Denmark, 2020.

Martinez, L. Development of tubular perovskite-fluorite oxygen transport membranes for biomass gasification .PhD Thesis, DTU, 2020.

Martinez, L.; Puig Arnavat, M; Ovtar, S.; Bjørnetun Haugen, A.; Kaiser, A. Ahrenfeldt J.; Henriksen, U., Hendriksen, p.V.; Gurauskis, J.; Kiebach, W.R., Partial oxidation of biomass gasification tars with oxygen transport membranes, Manuscript to be submitted 2021.

Garcia Fayos, J., Martinez L.; Bjørnetun Haugen, A.; W.R., Serra, M.J., . Y0.8Ca0.2Cr0.8Co0.2O3-δ – based dual phase membranes for O2 separation for oxy-fuel and syngas applications, Manuscript to be submitted 2021.

Martinez, L.; Bjørnetun Haugen, A.; Kaiser, A; Kiebach W.R., Interaction of Y0.8Ca0.2Cr0.8Co0.2O3-δ – Ce0.9Gd0.1O2-δ dual phase oxygen transport membranes on tubular Zr0.97Y0.6O2-δ, manuscript to be submitted. 2021.

"Danish roadmap for sustainable gas grid transition – status of the role of thermal gasification" https://orbit.dtu.dk/en/publications/danish-roadmap-for-a-sustainable-gas-grid-transition-status-and-p

Gantt diagram

Based on Energinet.dk template doc. No. 15/05241-10

Project title: Highly Flexible Energy Production by Oxy-Fired Biomass Gasification – Switching between Electricity, Gas and Liquid Fuel Generation (HighFlex)

Project starts: 01.09.2016 (updated project end after prolongation: 31.07.2020)

Work packages:	2016												2017												2018												2019												2020											
	1	2	3	4	5	6	7	8	9	10	11	12	1	2	3	4	5	6	7	8	9	10	11	12	1	2	3	4	5	6	7	8	9	10	11	12	1	2	3	4	5	6	7	8	9	10	11	12	1	2	3	4	5	6	7	8	9	10	11	12
WP1 Membrane development									▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓															
WT1.1 Optimization of membrane geometry, materials and ceramic processing route									░	░	░	░	░	░	░	░	░	░	░				░																																										
WT1.2 Up-scaling and improving reproducibility of manufacturing																		░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░															
WP2 Demonstration of long-term stability and mechanical robustness									▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓														
WT2.1 Long-term testing under gasifier relevant conditions																				░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░														
WT2.2 Testing of mechanical reliability under relevant conditions														░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░																						
WP3 Integration and demonstration of OTMs in biomass gasification processes																						▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓						
WT3.1 Membrane testing for partial oxidation of tars																						░	░	░	░	░	░	░																																
WT3.2 Integration of the membranes and oxygen-blown operation of the gasifier																																░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░						
WP4 Techno-economic assessment of oxygen blown biomass gasification																	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓	▓					
WT4.1 Techno-economic analysis of integration of membranes in different biomass gasifier designs																										░	░	░	░	░	░	░	░	░	░																									
WT4.2 Simulation of integration of oxygen blown biomass gasification into the Danish energy grid																																	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░	░					
Milestones																																																												
MS 1.1.1 Dip coating slurries and sintering program for all functional layers developed																				🟩																																								
MS 1.1.1 Optimal component geometry selected for two stage gasifier, and membrane supports of that geometry extruded and characterized.																										🟩																																		
MS 1.2.1 Ceramic materials supplied to DTU by CoorsTek.									🟩																																																			
MS 1.2.2 Analysis of membrane fabrication at DTU performed																							⬛																					🟩																
MS 1.2.3 Membranes with a total of 4000 cm2 of active membrane area with reproducible properties fabricated																																								|🟩|																			
MS 2.1.1 Rig for long-term-testing constructed and commissioned after internal safety approval																				🟩																																								
MS 2.1.2 Membrane tested for 5.000 h according to defined testing protocol																																|🟩|																											
MS 2.1.3 Membrane (from identical production batch used in MS 2.1.2) tested for 8.500 h																																								|🟩|																			
MS 2.1.4 Post mortem analysis on membrane tested in MS 2.1.2 performed and compared with results from non-tested sister membranes. Report on degradation mechanisms issued																																			|🟩|																								
MS 2.2.1 Testing protocol for mechanical testing based on input from all partners established																				🟩																																								
MS 2.2.2 Ceramic membranes developed in WP1 tested according to the protocol defined in MS 2.3.1																																						|🟩|																					
MS 3.1.1 Membrane performance for partial oxidation of tars determined																												|🟩|																															
M3.1.2 Best position for membrane integration in the TwoStage gasifier determined																														|🟩|																													
MS 3.2.1 Successful integration of a membrane tube module and proof-of-concept run with oxygen blown biomass gasification with the TwoStage gasifier at DTU																																																							🟧	→	→	→	→	⬛	
M3.2.2 Recommendations and guidelines for a >100kW oxygen blown biomass gasifier design																																																							🟧	→	→	→	→	⬛	
MS 4.1.1 Calculation of economic figures of merit for membrane integration in different integration schemes																																	|🟩|																										
MS 4.2.1 Danish road map on optimal integration of oxygen blown biomass gasification including economic evaluation criteria and influence on the grid stability provided to public																																																					🟧	→	→	→	→	⬛			

status 12.08.2019

🟩 full-filled mile stone

🟧 mile stone effected by project prolongation

⬛ mile stone to be full-filled in the future