DELIVERABLE REPORT

ECo - Efficient Co-Electrolyser for Efficient Renewable Energy Storage

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Deliverable summary

The overall goal of ECo is to develop and validate a highly efficient co-electrolysis process for conversion of excess renewable electricity into distributable and storable hydrocarbons via simultaneous electrolysis of steam and CO_2 through SOEC (Solid Oxide Electrolysis Cells). It is expected that the ECo project will have major environmental, social, economic, and technological impacts.

Already in the first period of the project, it was shown how the ECo project concept can contribute to Europe's' targets of reducing greenhouse gas emissions, of increasing the share of renewable energy sources in the energy mix, and of reducing the primary energy use – both as conceptual studies with specific cases and also by system design studies. Economic benefits are currently under detailed investigation for specific cases including the capture of CO_2 from different sources. The technological impact is already evident from the development and demonstration of improved versions of solid oxide electrolysis cells. More benefit is expected regarding durability on cell and stack level as compared to state of the art.

More specifically, the following results in relation to the objectives of the ECo projects were achieved:

- An improved SOEC was developed through electrode optimisation, which allows for operating the SOEC at ~50-100 ^{o}C lower temperature as compared to the current state. This achievement needs to be confirmed for relevant cell sizes and also in a stack environment.
- Durability tests have been carried out under realistic co-electrolysis operating conditions on state of the art cells and stacks in order to benchmark the technology state. The degradation rates are in a range, which is not sufficient for commercial use. The obtained results represent important knowledge to base further durability studies on. Improved cells and stacks will be studied for expected better durability in the forthcoming project period.
- An SOEC plant was designed that lays the basis for evaluating the impact of operating parameters on methane production rate and system efficiency. In the coming period, more details will be implemented such as the direct internal methane formation inside the SOEC stack in order to support the general project concept and to guide the selection of operating conditions for the final system test in the ECo project.
- Realistic cases for the ECo project were selected, which include all needed sources for the SOEC process (gasses and electricity) and which currently serve as basis for the evaluation of economic viability and also for the life cycle assessment.

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Background & Objectives of the ECo project

Europe has ambitious climate goals in order to improve environmental conditions and counteract global warming. The reduction of greenhouse gas emissions and the increase of energy production from renewable sources are on the agenda. Particularly the establishment of more and more wind and solar based electricity production leads to challenges because of the fluctuating nature and not predictable pattern of these renewable sources. Efficient technologies for storage – also on large scale, distribution and balancing are needed.

High temperature electrolysis provides a solution for these challenges. Using solid oxide electrolysis cells (SOECs), steam and even CO_2 can be directly converted in an electrolysis process that can use excess electricity from renewable sources. This process is called co-electrolysis and is unique for the SOEC technology. The electrolysis efficiencies are very high, around 90%. The obtained gas mixture is known as synthesis gas (hydrogen + CO) and can be converted by known catalysis technology into hydrocarbons such as methane or methanol. Methane is in fact a very attractive storage and distribution medium because an extensive natural gas network and storage tanks exist throughout the whole Europe. Already now, approx. 50% of the total electricity produced from renewable sources could be accommodated as methane in existing underground storage facilities. Thus no additional infrastructure is necessary, saving significant investments. The comprehensive natural gas network also makes it a system of transport for bringing gas from production to consumption areas with minimal losses.

The overall goal of ECo is to develop and validate a highly efficient co-electrolysis process for conversion of excess renewable electricity into distributable and storable hydrocarbons via simultaneous electrolysis of steam and CO_2 through SOEC (Solid Oxide Electrolysis Cells). The project approach is illustrated in Figure 1.

Figure 1. ECo approach of using electricity from fluctuating sources like wind and solar, together with steam, and CO_2 from sources like biogas or the cement industry in the SOEC process, yielding synthesis gas, which is converted in a catalytic unit to hydrocarbons like methane, which in turn can be added to the existing natural gas infra structure

More specifically, the ECo project will:

- Improve the current SOECs to achieve high performances and high efficiencies at ca. hundred degrees lower operating temperatures than state of the art in order to reduce thermally activated degradation processes, to improve integration with hydrocarbon production, and to reduce overall costs.
- Investigate durability under realistic co-electrolysis operating conditions that include dynamic electricity input from fluctuating sources and high pressure operation in order to reduce costs for the integration of SOEC with the subsequent catalytic processes and gas distribution. The aim is to acquire unique knowledge about durability and to finally achieve low degradation rates under such relevant operating conditions.

- Design a plant to integrate the co-electrolysis with fluctuating electricity input and catalytic processes for hydrocarbon production, with special emphasis on methane production and perform selected validation tests under the thus needed operating conditions.
- Test a co-electrolysis system under realistic conditions for final validation of the obtained results at larger scale.
- Demonstrate economic viability for overall process efficiencies exceeding 60% by identifying and evaluating concept designs based on real cases.
- Perform a life cycle assessment with CO_2 from different sources, which are realistic in future, fossil free energy systems (like cement industry or biogas) and electricity from renewable sources.

It is expected that the ECo project will have major environmental, social, economic, and technological impacts, particularly because it investigates SOEC under realistic conditions, improves the current SOEC, and projects the technology towards real cases. At the end of the ECo project, a higher technology readiness level than state of the art will be achieved, thereby moving the technology closer to a market break through.

Organisation of the Project

The project is organised in three technical work packages that are aimed at addressing the issues as outlined in the Work Plan, one management WP and one for dissemination/exploitation (Figure 2).

In WP1 (SOEC Development), improved cells will be developed addressing known sources for degradation of SoA cells through structure and design development. Improved electrodes will be integrated into full cells, that allow for the reduction of operation temperature by 100 ^{o}C compared to SoA cells, and stacks and the improvement of performance and durability will be verified through testing under selected conditions in WP2 compared to SoA cells.

In WP2 (SOEC Co-electrolysis Process), operation at relevant environment at cell and stack level will be investigated with the aim to identify limiting conditions and optimum conditions to achieve below 1% degradation/ 1000 h. Relevant environment will include effects of temperature, pressure (for better integration with fuel production), dynamic operation (to simulate fluctuating electricity from renewables), and different H_2/CO ratios (to prepare for different downstream hydrocarbon production). The testing will also include internal methanation as ultimate degree of integration of electrolysis and catalysis to achieve highest possible efficiencies. Obtained test results will be used for further cell improvement in WP1 and for analysis in WP3. Test at system level will be performed applying the optimum conditions as identified from comprehensive test and from techno-economic analysis in WP3.

In WP3 (SOEC Analysis), a system will be designed based on the results obtained in WP2. A techno-economic analysis will be performed including a competition analysis to SoA technologies for synfuel and methane production. Results on improved cells from WP1 will be included to give a prognosis on needed further improvements and what key parameters that have to be achieved for a cost competitive co-electrolysis technology. Further, a life cycle analysis will be performed including CO_2 input from different sources – taken into account sources that will persist also in a fossil-free energy system.

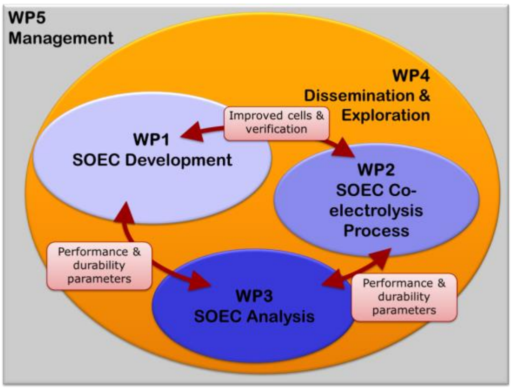

Figure 2. Illustration of project organisation

Results of Work Package 1: SOEC Development

Objectives of WP1

The main objectives of WP1 were to develop and optimise SOEC cells for co-electrolysis based on the fuel electrode supported cell concept in order to achieve an improved performance, which allows a decrease of the operating temperature by 50-100 degrees (from ~800 to ~700 ^oC) while keeping the same performance as SoA cells. Improved electrodes were to be integrated into SoA cells and improved full cells into stacks for verification tests in WP2. These objectives were approached by modification/ optimisation of both, the oxygen and the fuel electrode and by:

- Applying a new backbone/infiltration concept (DTU, IREC, see illustrations in Figure 3)
- Structural optimisation of the oxygen electrode by depositing several thin layers of LSCF/GDC composites, with different mixtures and structural features based on results of 3D reconstruction and associated modelling (CEA, see Figure 3).
- Improvement of the Ni/YSZ fuel electrode to suppress one significant degradation mechanism related to the loss of Ni percolation at the electrolyte/electrode interface, by optimisation of Ni particle size, content and particle size distribution and pores sizes (DTU).

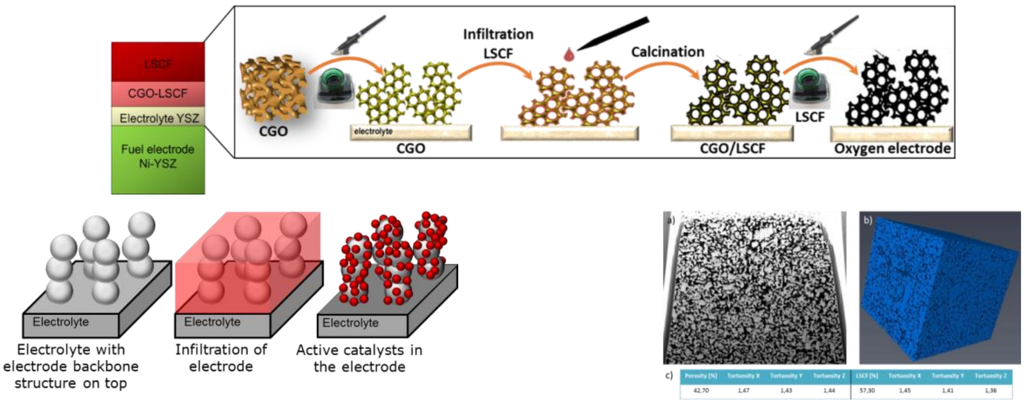

Figure 3 Scheme of the fabrication process of the functional active layer on the oxygen electrode side of the IREC (top) and DTU (bottom, left) SOECs and a) SEM micrograph, b) 3D reconstruction and c) microstructural parameters measured with SoA CEA electrode (bottom, right).

Summary of WP1

Two versions of improved cells were developed so far. The first version is based on the optimisation of the micro structure of the LSCF/CGO oxygen electrode aided by detailed electrochemical studies, modelling, and 3D micro structural analysis. The second improved version is based on an optimised structure and composition of the Ni/YSZ fuel electrode. Future candidates for improved cells comprise oxygen electrodes with high surface area backbone structures, where the mixed electronic ionic conductor is infiltrated.

The improved cells were delivered to WP2 for durability testing and to HTc for integration into stacks.

The activities in WP1 have been very successful in terms of yielding improved cells. Particularly, the close collaboration between the partners has to be mentioned, where cells and cell components were exchanged among each other, leading to improved cells that contain combined know how and optimised layers from more than one partner.

The improved cell version 1 developed by CEA shows a ca. 20% better initial performance than the SoA HTc cell, which allows operating this cell at a ca. 100 ^{o}C lower temperature. This achievement corresponds to the first objective of the project and needs to be confirmed at the large cell and stack level in the coming project period.

Results of Work package 2: SOEC Co-electrolysis Process

Objectives of WP2

The objectives of WP2 were to test SoA and improved cells and stacks under realistic operating conditions, including test at atmospheric and high pressure, dynamic conditions, comparison between co-electrolysis and steam electrolysis mode, and potentiostatic tests. In order to obtain better understanding, the electrochemical tests are to be coupled with micro structural analysis and modelling. For a better comparison of results between the partners, a harmonised test protocol has to be established.

Impurities in the CO_2 from relevant sources may impact the performances of SoA cells. Such CO2 product streams will be analysed and the impact of selected impurities on the performance and durability of SoA cells will be tested.

In the final project phase, a system test under optimum conditions yielding degradation rates below 1% efficiency per 1,000 h lifetime will be carried out

Summary of WP2

In the initial phase of the project, a comprehensive test protocol was defined with the aim to guide the testing of cells and stacks at the partners' labs in order to utilise the existing test facilities in a most efficient way to be able to cover the promised large operating parameter space. The test schemes consist of a sequence of gas settings, current-voltage (i-V), electrochemical impedance (EIS) measurements and cell operation under electrical load, including ranges for variation of operating parameters like temperature, inlet gas composition, pressure etc. It was based on previous European projects like SOCTESQA, RelHy, and ADEL. It has to be stressed that a standardised testing under relevant SOEC conditions is technically very challenging and is still a long process before it is achieved. In the ECo project, a higher level of harmonisation of testing will be achieved and it will be possible to utilise the testing facilities at the testing partners in an efficient way because results are – with certain limitations – comparable.

The SOEC testing partners (EPFL, CEA, DTU, EIFER) tested SoA cells delivered by HTc for initial performance and to a certain extent for durability at selected conditions (see Table 1). As they became available, improved cells were included in the testing. The detailed assessment carried out so far has led to new knowledge about the performance and durability of SOEC under realistic conditions, which can be used in WP3 for modelling of the SOEC system. Generally, the performance of the cells decreased (i.e., the ASR increased) when going from a steam/hydrogen gas mixture to co-electrolysis gas mixtures containing more and more CO_2. The reason for this increase of ASR is mainly due to the increase of the low frequency part of the impedance, which is related to gas transport and conversion.

Table 1 Performance test results of HTc SoA cells at the different partners; the current density at 1.3 V and an estimated reactant utilisation close to 50-60% at -1 A/cm² was selected as performance parameter at 750 °C and a gas mixture of 25% CO_2/65% H_2O/10% H_2

Partner	Cell	I (A/cm^2) at 1.3 V
EPFL	Circular, diameter 60 mm, active area 12.56 cm^2	0.97 @ RU= 62%
DTU	Square, 5 x 5 cm^2, active area 16 cm^2	1.01
EIFER	Square, 5 x 5 cm^2	0.85
CEA	Circular, active area=3.14 cm^2	0.97* @ RU= 62%

* Average of three tests 0.97 ±0.3 A/cm²

The durability tests showed clearly that running cells and short stacks under relevant co-electrolysis conditions over hundreds to thousands of hours is still a challenge, both technical and also cell/stack related. Particularly, the steam supply seems to be a critical issue and the shut-down procedures in case of emergencies such as the steam supply failure. It is very important for the partners to collect such experiences and to learn from the encountered problems. Nevertheless, degradation data of SoA cells and stacks were obtained with cell voltage degradation rates in the range of few to tens of %/kh. The results show, that the durability of SoA cells and stacks needs to be improved, which is one of the objectives of the ECo project.

It was observed that the performance of the cells in a short stack environment was slightly reduced compared to single cells, indicating extra resistance contributions from stack components and/or stack design. On the other hand, the degradation rates in co-electrolysis mode seem to be in the same range or even better, i.e. there seem to arise no additional degradation processes from stack components and the cell is the main source of degradation in the SOEC process.

Further analysis of the EIS of single cells and short stacks, for example using the distribution of relaxation time method (DRT), is currently carried out to explain the differences of initial performance and the differences of the degradation mechanisms are expected to be identified. This analysis will be combined with scanning electron microscopy (SEM) combined with energy diffraction spectroscopy (EDS).

Results of Work package 3: Technical-economic analysis and life-cycle assessment

Objectives of WP3

In WP3 a solid oxide electrolyser system for the production of hydrogen or syngas for process coupling with downstream catalytic reactors for gas synthesis (methanation) or internal methanation will be designed and analysed. This includes:

- Process concepts with flow sheeting of power-to-gas and power-to-liquid plants with high temperature electrolysis taking into account feedstocks (H_2O, CO_2 and renewable electricity) and products (purity, quantity etc.)
- Life cycle assessment of SOEC plants with special focus on CO_2 balance
- Techno-economic analysis of SOEC plants and competitiveness under various business models, benefits, life cycle assessment on the CO_2
- Specifications for a demonstrator taking into account technologies maturity and favourable applications for future business cases

Summary of WP3

Major tasks accomplished in WP3 include the modelling and model-based understanding of an SOE based Power-to-Methane (PtM, see Figure 4) system without CO_2 capture. It was found that there is a trade-off between the system efficiency and methane production rate. A better system-level heat integration is possible by co-electrolysis operation, which leads to higher efficiencies compared to steam electrolysis. An exothermic operation mode has the potential for capacity boost of a fixed hardware.

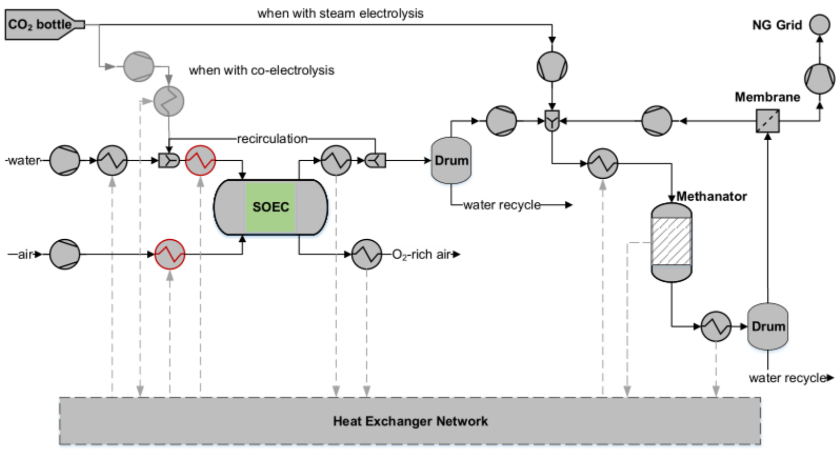

Figure 4 ***EPFL*** *Schematic of the considered SOE-based PtM system without explicit heat exchanger networks. The CO_2 feed streams are distinguished for both steam- and co-electrolysis, and the heaters in red may be partially driven by electrical to heat up the targeted steams to the required temperature. The components needed for start-up, hot stand-by and temporary gas storage are not included.*

A number of different power-to-gas/liquid concepts were investigated in terms of general features, efficiencies, electrical power consumption and investment costs at different sizes and for co- and steam electrolysis.

Realistic CO_2 sources were evaluated regarding CO_2 capture technologies and suitability for combination with the SOEC technology. In this process a comprehensive literature study was carried out and – maybe even more importantly – suppliers of CO_2 as present in the Industrial Advisory Board and as partner in the ECo project were involved.

Specific cases for the use of SOEC technology in combination with real CO_2 sources from cement industry and from biogas were identified and outlined for further economic and life cycle analysis.

Results of Work package 4: Dissemination & Exploitation

Objectives of WP4

The objectives of WP4 are:

- to coordinate and propose means of dissemination to different communities (scientific, industrial, general public, public bodies, etc.)
- to provide public technical data on project activities and results of SOEC development
- to build an exploitation strategy identifying and managing exploitation opportunities
- to manage and monitor IPR related issues

Summary of WP4

Communication and image tools where created in order to consolidate the corporate image of the ECo project. There has been a lively dissemination of the project concept and the first project results to a broad community, including the public in general, the industry and the scientific community. Many different media have been used for this dissemination, public events, visits, poster and oral presentations, website, brochure, etc. In that way, the communication has reached a large variety of audience at different knowledge and influential levels, such as for example politicians, school kids, the general public, university students, the science community, and industrial players. In the coming project period more focus will be on the exploitation of the achievements.

Results of Work package 5 Management

Objectives of WP5

The overall objectives of WP5 are to setup and execute the project management tools and bodies, such as:

- Handling of all administrative, financial and legal issues in the project to guarantee contractual engagement and interface with the FCH-JU
- Monitoring of work progress, difficulties and reacting to major problems to achieve the project goals
- Ensuring optimal internal and external communication to achieve good interaction between work packages, partners and external entities
- Scheduling and organising of efficient project meetings
- Central financial administration, monitoring and control.

Summary of WP5

The management of the project has been very successful in general. The management structures and bodies worked well and efficient in order to support the progress of the project, monitor the activities, discuss challenges and suggest solutions to those. The consortium has been very motivated and engaged to achieve the targets set in the project. Regular physical (rotating between the partner sites, see Figure 5) and phone meetings were held to report on progress and status in general.

Some delays of reporting in the first phase of the project were due to the complexity of the project and that partners had to achieve sufficient degree of exchange of information and knowledge.

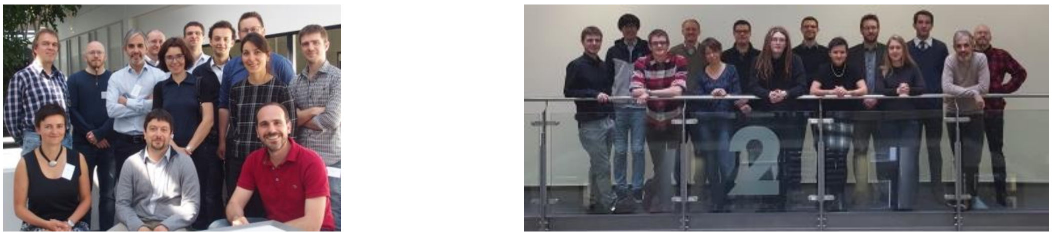

Figure 5 Pictures from the 1st (kick-off at DTU in Risø/Copenhagen, Denmark) and so far latest (3rd half year at EIFER in Karlsruhe, Germany) meetings

Exploitation of results

In the first project period, a significant progress in the field, in relation to new SOEC knowledge, new cells and stacks, and system designs have been developed, which are interesting for commercial exploitation. In the coming period, there will be more focus on these exploitation issues with examples listed in Table 2.

Table 2 List of results with potential for exploitation and stakeholders

Achievement	Stakeholders
Improved SOEC cell based on fuel electrode supported concept	DTU, CEA, IREC, HTc
Definition of a harmonised testing protocol based on previous European projects	JRC, SOEC testing community
Design of SOEC plant	HTc (Solid Power), Sunfire
Definition of application cases and techno-economic analysis	VDZ, Enagas, Engie/Laborelec, HTc/SolidPower