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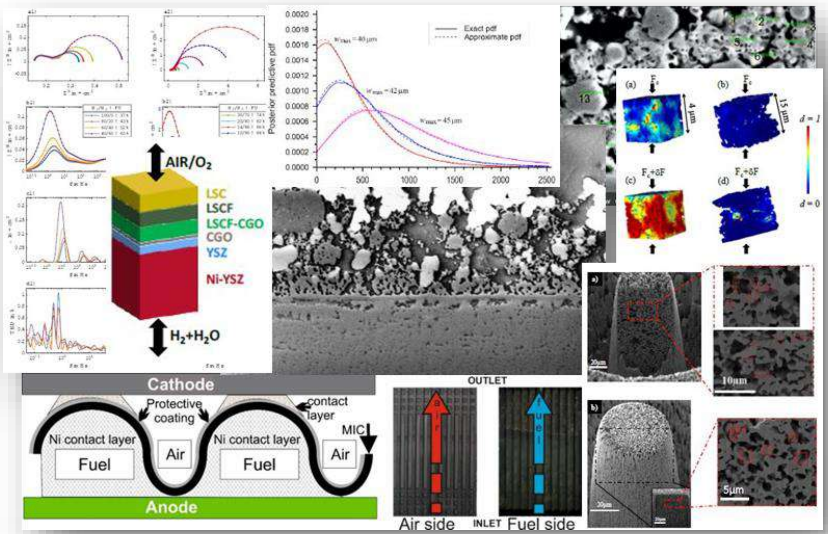
Clean Hydrogen Partnership

HADASTRA

"HArnessing Degradation mechanisms to prescribe Accelerated Stress Tests for the Realization of SOC lifetime predictive algorithms".



Accelerated stress test procedures for solid oxide cells: Pooling recognized excellence in industrial practice and scientific know-how towards the realization of validated protocols

Layman's Report 2022

AD ASTRA – "HArnessing Degradation mechanisms to prescribe Accelerated Stress Tests for the Realization of SOC lifetime predictive algorithms". Accelerated stress test procedures for solid oxide cells: Pooling recognized excellence in industrial practice and scientific know-how towards the realization of validated AST protocols

Final Layman's project report for the European project "HArnessing Degradation mechanisms to prescribe Accelerated Stress Tests for the Realization of SOC lifetime predictive algorithms (AD ASTRA)", funded under the H2020 Programme of the European Union, supported by the Clean Hydrogen partnership under Grant Agreement: 825027

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1 PUSHING THE LIMITS OF INNOVATION

The game-changing progress that has been achieved in the last decade in terms of sustainable technologies, resource diversification and electrification is leading the world to attempt a bunny hop into full defossilisation. The contribution of innovation in all aspects of human life has hardly ever been more prominent and this is pushing the boundaries of what was thought possible in terms of energy supply, value chain governance and technological capability. In this panorama, harnessing the physical and chemical phenomena that determine production processes, predicting and controlling them, has become the new paradigm.

For the feasibility and sustainability of the energy transition from fossil fuels to renewable one, the losses in the energy conversion chain need to be minimised and material life cycles need to be maximised. Increasing the flexibility of the energy system requires seamless sector coupling and a tapestry of complementary technologies that operate at maximum efficiency with real-time interconnectivity and full cascading of by-products and auxiliary flows. Electrochemical systems are the new frontier in this respect, and have the potential to become a crucial technology in powering the transition to clean and efficient energy.

What Solid Oxide Cells – SOC – can do

Among the different electrolysis technologies, the Solid Oxide Cell (SOC) is attractive both because of unrivalled global conversion efficiency - over 90% (electrical + thermal) - and because it can operate in reversible mode, switching between hydrogen production mode or power storage (SOEC, Solid Oxide Electrolyser Cell) and power generation (Solid Oxide Fuel Cell, SOFC) as required. These properties make them a technology hub for hydrogen production, distribution, and consumption, simultaneously balancing the electricity grid. A schematic representation of an r-SOC system for the balancing of the electricity grid involving Renewable Energy Systems (RES) is depicted in Figure 1.

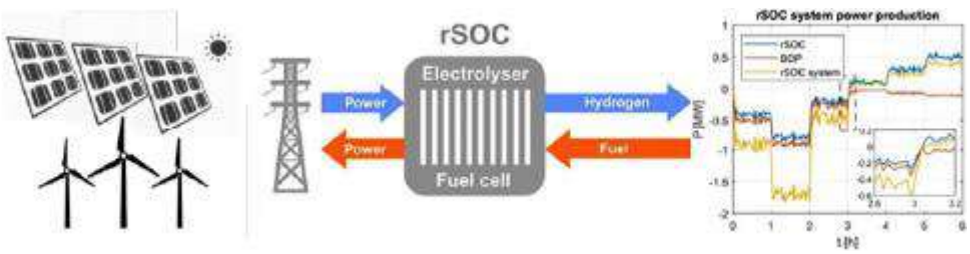

Figure 1: Simple schematic of an r-SOC system for RES balancing.

Solid oxide technology has undergone tremendous development and improvement over the last decades. This has led to the growth of various industrial realities throughout Europe, ranging from individual cells development and production to the construction and commercialization of small-sized modules (a few kW) up to modules of hundreds of kW. Despite these advances, research and development challenges remain, tied to necessary cost reduction and lifespan enhancement, before wide deployment can be achieved. In this context, the **AD ASTRA** project aims to develop **Accelerated Stress Test (AST)** protocols that allow quantitative identification and prediction of critical degradation mechanisms, correlating them with overall performance variables in selected SOC stack components (fuel electrode, oxygen electrode and interconnect), leading to improvement of SOC stack durability, savings in stack acceptance efforts, shorter time-to-product cycles of innovative SOC concepts, a methodological framework for resource-efficient certification of SOC stacks.

2 SOC BASIC PRINCIPLES

The SOC is an electrochemical conversion system based on the Solid Oxide Cell technology which can be reversibly operated between fuel cell (SOFC) and electrolyser (SOEC) modes. Its electrodes can be effectively operated in both operating modes, thanks to the compatibility of the electrocatalysts with both electrochemical conversion directions. The high operating temperatures (around 800°C) favour the reaction kinetics and ensure that all reactants are always in gaseous phase: water is therefore produced/consumed in the form of steam, which demarks its most feasible applications where this is needed or available. By implementing a suitable balance of plant, the system can be unitized in a single poly-generation system which can act as a flexible energy storage system switching between SOEC mode, hence generating hydrogen from steam and electricity, and SOFC mode, hence generating electricity and steam from hydrogen. In both operating modes, a high-grade heat recovery is possible thanks to the high operating temperatures of the process. A schematic representation of each operating mode for SOC systems is reported in Figure 2.

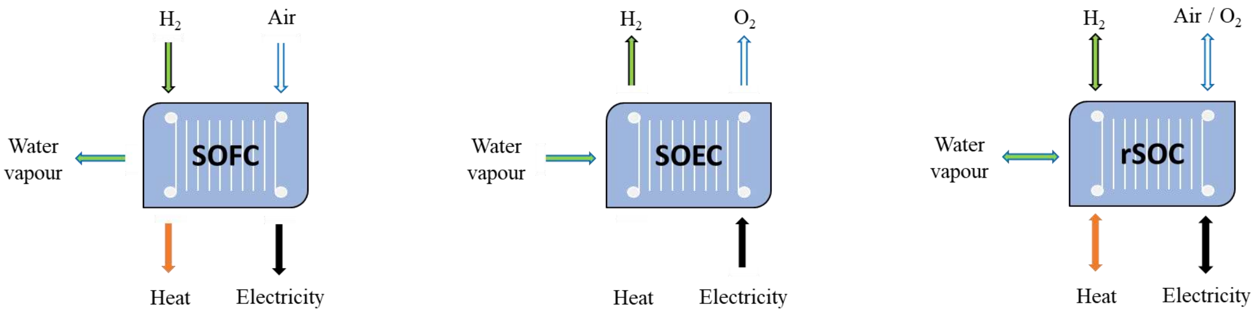

Figure 2: Schematic representation of (a) SOFC system; (b) SOEC system; (c) rSOC system.

SOFC mode

Figure 3 displays the building block of the SOFC: each of these cells – consisting of an anode, electrolyte and cathode – can be connected and stacked up to provide any requirement of power. This modular build-up is what makes it possible for the SOFC to have practically constant efficiencies from Megawatt to single watt scale. The fuel is fed to the negative electrode, where the high temperature allows it to be separated into its essential constituents. In hydrocarbons, these are hydrogen (H_2) and carbon monoxide (CO). H_2 and CO react in the same way at the fuel side. Taking H_2 as an example, it reacts electrochemically to generate two electrons per molecule of hydrogen. This current is made to flow across the electrical load that needs to be powered and reacts at the positive electrode with the air – or the oxygen (O_2) in particular – that is provided there. Every two electrons generate an oxygen ion (O^{2-}), which migrates across the gas-tight electrolyte to the fuel electrode, where it reacts with the hydrogen to release again the two electrons that generated the O^{2-} ion, effectively closing the electrical circuit. In the process, the only by-product formed is water. In the case of CO, the by-product is CO_2. Therefore, the outlet of the SOFC produces a clean and relatively pure mixture of water and carbon dioxide. Thus, if necessary, the carbon dioxide can be separated and sequestered much more easily than is the case with the by-product flows from combustion, where large quantities of nitrogen, contained in the air used for combustion, dilute the CO_2 content and make it energy- and cost-intensive to separate. Furthermore, the potential to generate clean water could make them attractive for areas and applications where water is in short supply.

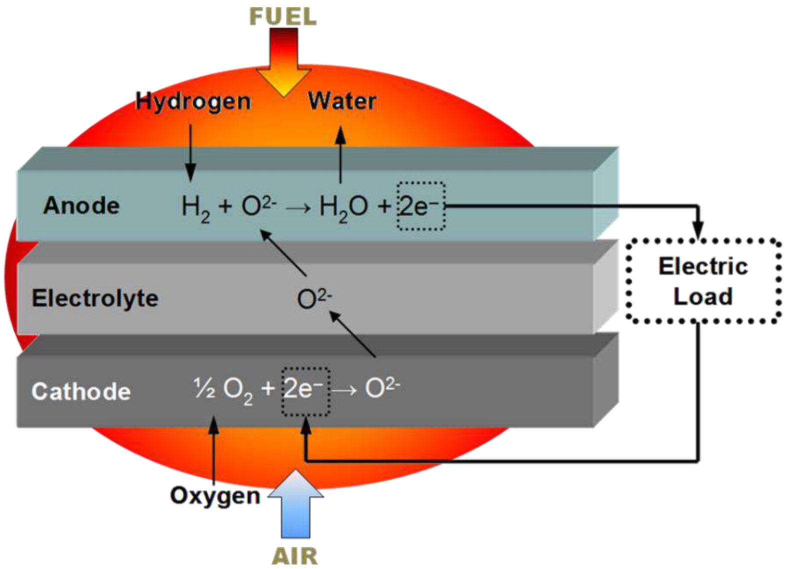

Figure 3: How the SOFC generates high-efficiency power and heat from fuel and air

To turn the stack of cells to a fully functional power generating system several auxiliary components (the so-called balance-of-plant, BoP) have to be integrated, taking care of fuel pre-treatment, power management and heat exchange. In order to preserve the high efficiency of electrochemical conversion in the SOFC, the BOP often needs to be designed and produced specifically to optimize the integration and minimize parasitic losses. This is an important part of turning the SOFC to real, viable end-products.

SOEC mode

The key components of a SOEC are the same as of a SOFC: a dense ionic conducting electrolyte and two porous electrodes. The fundamental mechanisms involved in SOEC operation are shown in Figure 4a. Steam is fed to the porous fuel electrode. When required electrical potential is applied to the SOEC, water molecules diffuse to the reaction sites and are dissociated to form hydrogen gas and oxygen ions at the fuel electrode–electrolyte interface. The produced hydrogen gas diffuses to the electrode surface and gets collected. The oxygen ions are transported through the dense electrolyte to the anode. On the anode side, the oxygen ions are oxidized to oxygen gas and the produced oxygen is transported through the pores of the anode to the anode surface.

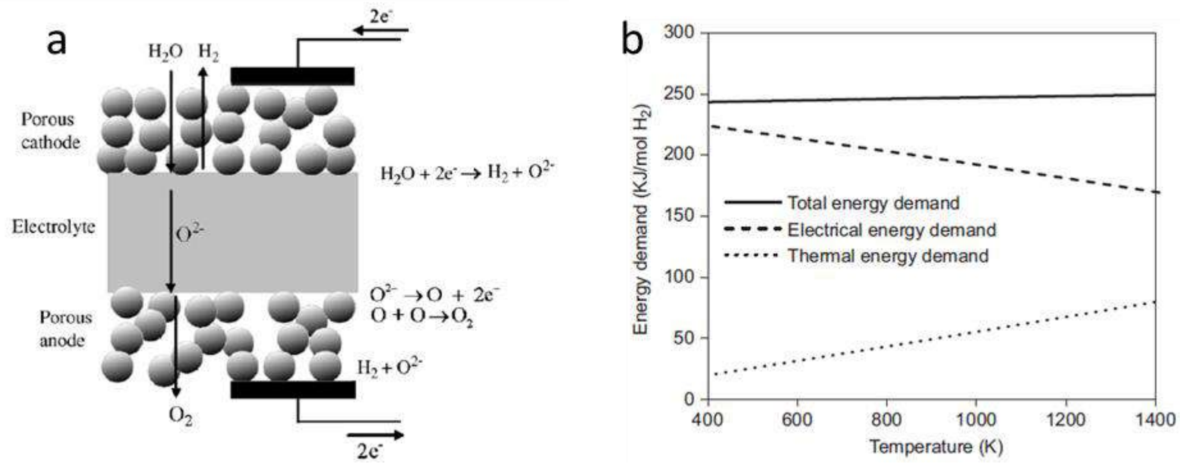

Figure 4: a) Working principle of SOEC; b) Calculated energy demands for electrolytic H_2 with varying temperatures.

Figure 4b shows the calculated energy demands with varying temperatures. An increase in operating temperature decreases the electrical energy demand and increases the thermal energy resulting in a lower operating voltage (1.2 to 1.4 V). The lower operating voltage of the SOEC enables hydrogen production with less electrical energy consumption in its stack. As the greatest portion of the hydrogen production cost through electrolysis is the electricity cost, these thermodynamic benefits of the SOEC directly lead to an economic advantage, represented by its cheaper hydrogen production cost (in €/kgH_2).

3 THE AD ASTRA PROJECT

AD ASTRA has been a collaborative project to address a scientific challenge of global importance that will lead to the development of concrete tools for industries producing SOC components and stacks. Industries in this field have so far not been able to divert resources from their core concern of cutting costs in manufacturing a standard product to the definition of reliable accelerated stress test (AST) protocols, that would shorten the time to product of innovative, more robust, and cost-effective technology. The project received 3-million-euro funding from the clean hydrogen partnership (ex FCH-JU) in support of the expected costs of the project. It started on the 1st of January 2019 and lasted until the 31st of August 2022.

The AD ASTRA consortium

The consortium (Figure 5) is formed on the basis of technical and scientific excellence and the ability to collaborate effectively, which have been shown by their long-standing bilateral and multi-lateral cooperation in previous and on-going projects. It can be asserted that many of the leading European research organizations and industries in SOC development and deployment are gathered in this project, each with tens of years of track record on the pathway to market readiness of the technology. The laboratories taking part in AD ASTRA have a record of quality assurance in SOFC/SOEC testing, modelling, and post-test characterisation, particularly through their joint participation in SOCTESQA (ENEA, CEA, DTU, EIFER) – that guarantees reliability and repeatability of the test results. Furthermore, the specific competences of each partner synergize perfectly:

· the two industries, Sunfire and SOLIDpower, develop SOC stacks that are of different architecture (electrolyte-supported vs. electrode-supported) and temperature (750 vs. 850°C) and each focuses on a different application area (CHP and P2X), and have a large body of field tested stacks available for the project

· CEA, EIFER and ENEA are among the leading energy research centres in Europe, with a wide range of important infrastructures for large-scale technology transfer, as well as benefiting from national mandates for policy support and standardization of technology and procedures

· DTU, EPFL, UNIGE are top-level universities with longstanding direct collaborations with European fuel cell industries (Topsoe, SolydEra and Ansaldo Fuel Cells) giving them the empowering know-how to bring scientific concept to market

· IEES and UNISA are the entities that bring in highly specialized knowhow from adjacent scientific fields, namely electrochemical analysis, energy system modelling and control, and applied probabilistic mathematics, with a proven record of application in fuel cells technology

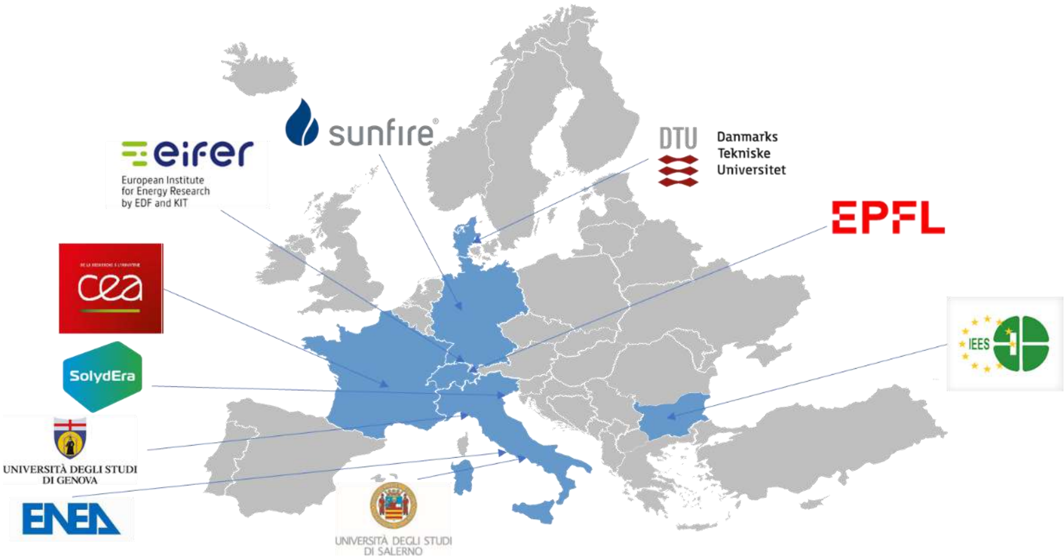

Figure 5: the AD ASTRA consortium

4 WHAT WE ACHIEVED

Characterization of field-tested samples and stacks

Designing and planning accelerated tests is based on two main pillars: to know how the operating parameters (i.e., temperature, pressure, gas composition, polarization) affect the degradation rate of the device components, and to identify the evidence of ageing in the materials as a function of the operating time. It is then possible to stress one or more operating parameters to accelerate the degradation process in a conscious way to help the prediction of materials behaviour during their operative life.

Based on these assumptions the starting point of the AD ASTRA project was to shed light on the field-tested stack samples provided by the manufacturers of the consortium. This was done by an extensive and structured experimental post-mortem analysis and subsequently applying the same methodology to the (accelerated) tested samples during the project aiming to verify the accuracy of the AST procedures elaborated by the consortium. Examples of characterisation results are depicted in Figure 6.

Thanks to this effort it was possible to acquire sufficient data to estimate the impact of the operating parameters on the acceleration of the degradation rate, creating a set of data to be used for comparison and cross check with samples issuing from the accelerated tests.

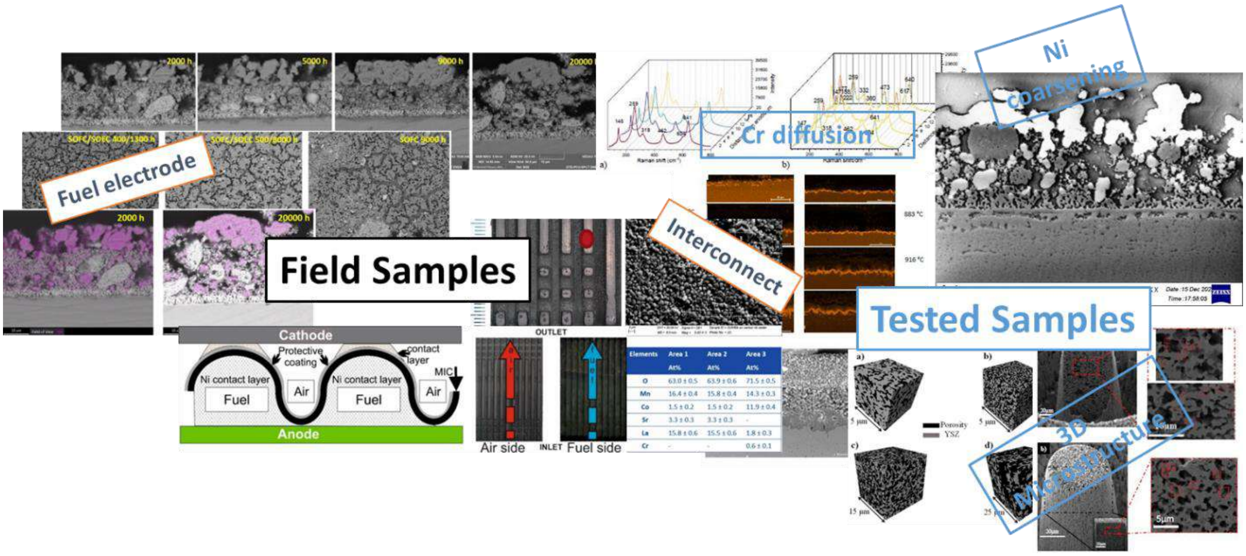

Figure 6: Results of post-mortem analysis on field samples operated in real world environment and tested samples in the project

Innovative testing approaches

Accelerated testing is a crucial element of an efficient design-to-product chain. Long-term reliability of durable systems cannot be feasibly proven in real-time tests, so that protocols need to be defined that characterize the behaviour of a given product towards end-of-life in much shorter times. For SOC, techniques like adding contaminants to the reactants or tripping by sudden reoxidation and thermal cycles have been attempted, but these approaches do not comply with rigorous scientific method nor fully satisfy the current industrial needs: applying stressors to the whole stack can over- or under accelerate irrelevant or relevant processes respectively, distorting the occurrence of degradation mechanisms and their compound effect on the stack performance. To systematically address, harness and accelerate the failure modes of the 3 components that are the focus of AD ASTRA, a matrix was set up of degradation mechanisms, test items, test procedures and characterization methods, that has been translated into a testing approach consisting of a dual-focus campaign targeting:

- **In-situ** aggravated tests of cells/stacks: these tests are designed for accelerated degradation phenomena caused by selected harsh values of an input operation parameter as temperature, load current, pressure, or extreme operation regimes as thermal or load cycling. The aggravated operational conditions must stress the sample in a representative way while minimizing time and resource expenditure.

- **Ex-situ** artificial ageing of critical cell/stack single components (fuel or oxygen electrode, interconnect) in appropriate accelerating conditions, which reproduce the aged state reached by calendar ageing in real life conditions. The level of ageing is determined based on analysis of the corresponding components extracted from a cell/stack after long term field tests. The artificially aged components can be integrated in an otherwise new cell/stack and further tested in-situ in the new configuration and nominal operating conditions, thus defining the effect of single component degradation on the total performance of the system. Another option is the artificial aging of the critical component in the final test assembly (cell, stack) prior to the further testing of the whole configuration. This approach (when possible) is more-simple and eliminates eventual influence of the final technological assembling on the already aged component.

Furthermore, a **radical innovation** consisted in the integration of these two methods: components specifically aged ex situ have been assembled into otherwise "new" stacks for in-situ testing, so that their effects on stack performance were easily isolated from other components that in real-world conditions would also degrade. This innovative approach led to the definition of a clearer identification and consequently acceleration of single degradation phenomena. Figure 7 summarizes some examples of stressor factors applied in IN SITU and EX SITU approaches.

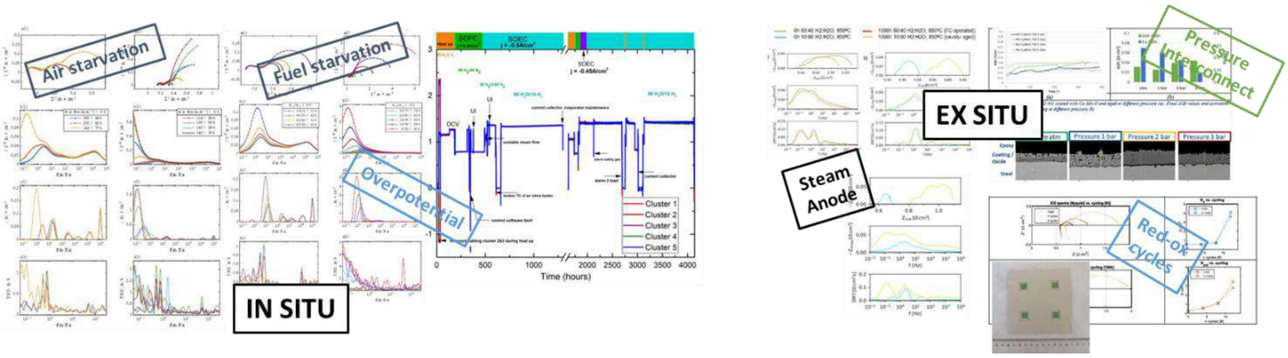

Figure 7: Examples of different stressor factors applied in IN SITU and EX SITU approaches

Remaining Useful Lifetime (RUL) predictive modelling and AST protocols design tool

Modelling activity had a central role in the project's studies on solid oxide cells allowing for a more concrete understanding of mechanisms and underlining the dominant correlations among several factors which influence the cell operation such as materials, geometry, microstructure and working conditions. Model results provide a useful guide to improve cell behaviour to make this technology competitive for long-lasting applications as both power plant and energy storage unit. In the framework of AD ASTRA project, different approaches have been proposed as performance and lifetime models to simulate how the cells behave during durability and accelerated stress tests. Indeed, both empirical and physics-based degradation functions have been introduced deriving them from experimental observations, specific high-level and grey-box degradation models.

Remaining Useful Lifetime (RUL) prediction during operation is important to organise maintenance intervals properly before a complete irreversible failure of cells or components. To this end, degradation-accelerating factors have also been correlated to cell operation through specific transfer functions that look at those parameters which mainly cause the worsening of performance and quantify their impact in terms of remaining lifetime as they occur. Such transfer functions can be then summarized as guidelines or design maps to improve AST protocols definition and application. An overview of the modelling approaches developed within the project is reported in Figure 8.

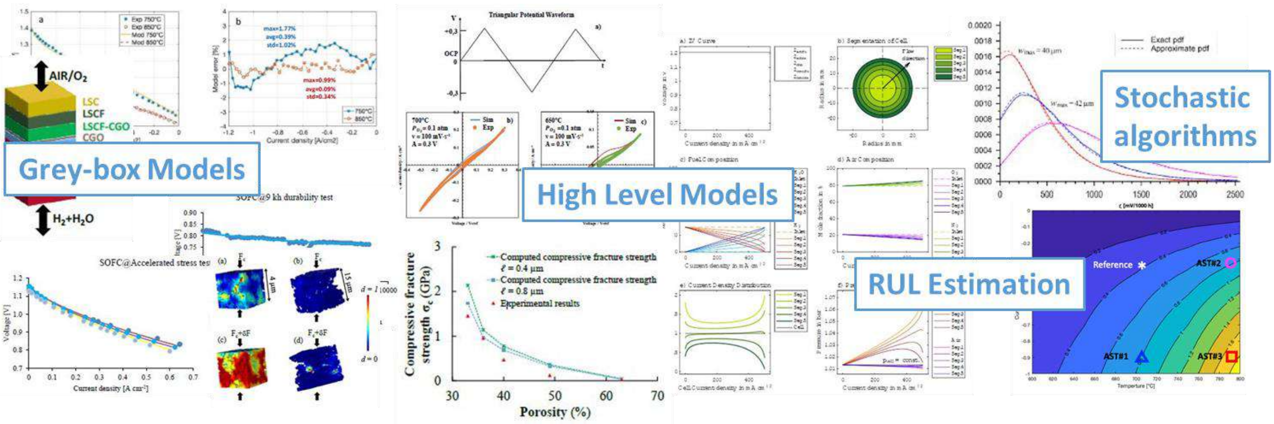

Figure 8: Modelling approaches employed and developed within the AD ASTRA project

AST protocols SOC for Strengthening the competitiveness of the European fuel cell industry

As a final result of the project 12 AST protocols were elaborated. They are divided in 2 main groups, reflecting the approach for the introduction of the stressor:

- In-situ aggravated tests for accelerated observation and testing of degradation phenomena in relation to test input parameters (marked in the title of the protocol) which generate harsh operational conditions (6 Protocols);

- Ex-situ artificial ageing (marked in the title of the protocol) which reproduces faster the degraded condition of a critical component or interface with respect to calendar ageing in nominal conditions (6 Protocols).

The protocols follow one and the same structure, giving information about: selected AST approach; samples used; experimental set-up; nominal operation parameters; stressor and stress factor; quantification of the degradation at nominal conditions and under stress test; acceleration factor.

Generally, all the results and particularly the definition of AST protocols helped significantly in increasing knowledge and understanding of degradation phenomena. Applying this knowledge will lead to a more focused improvement of the technology, helping to find new strategies for limiting the degradation of performances in long-term operation. The progress achieved by the project towards the definition of AST protocols, as well as in the definition of transfer functions and predictive algorithms for RUL estimation, will pave the way for the implementation of established procedures at industrial level capable of reducing resources and costs for the validation of the technology.

The activities of the AD ASTRA project, in conjunction with the project on PEMFC accelerated testing ID-FAST, have also given rise to a specific standardisation initiative on accelerated testing in the International Electrotechnical Commission's Technical Committee 105 (IEC TC105) on fuel cells, under Ad Hoc Group 11. The project has also established a liaison with the European Commission's Joint Research Centre (JRC) for the validation and harmonisation of the developed AST procedures within the compendium of European procedures for electrolyser testing.

Figure 9: The AD ASTRA team

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