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Advanced Electrolysers for Hydrogen Production with Renewable Energy Sources

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Abstract

The 3-year FCH project ADEL (ADvanced ELectrolyser for Hydrogen Production with Renewable Energy Sources) targets the development of cost-competitive, high energy efficient and sustainable hydrogen production based on renewable energy sources. A particular emphasis is given to the coupling flexibility with various available heat sources, allowing addressing both centralized and de-centralized hydrogen production market.

The ADEL 3-year-project target is to develop a new steam electrolyser concept, the Intermediate Temperature Steam Electrolysis (ITSE) aiming at optimizing the electrolyser life time by decreasing its operating temperature while maintaining satisfactory performance level and high energy efficiency at the level of the complete system, composed by the heat and power source and the electrolyser unit.

The project is built on a two scales parallel approach:

- At the stack level, the adaptation and improvement of current most innovative cells, interconnect/coating and sealing components for ITSE operation conditions aims at increasing the electrolyser lifetime by decreasing its degradation rate
- At the system level, to facilitate an exhaustive and quantified analysis of the integration of this "new generation ITSE" with different heat and power sources like wind, solar, geothermal and nuclear, flow sheets will be produced with adjustable parameters.

The paper presents data on electrochemical performance of specifically developed materials for electrolysis in a temperature range around 700°C. Conclusions of an international workshop are presented on where and under what conditions ITSE systems can contribute to the new, low-carbon energy system.

Introduction

The ADEL project (ADvanced ELectrolyser for Hydrogen Production with Renewable Energy Sources) proposes to develop a new steam electrolyser concept. This so-called Intermediate Temperature Steam Electrolysis (ITSE) aims at optimising the electrolyser life time by decreasing its operating temperature while maintaining satisfactory performance level and high energy efficiency at the level of the complete system including the heat and power sources and the electrolyser unit (Figure 1). The relevance of the ITSE is an increased coupling flexibility. Improved robustness and operability will be assessed both, at the stack level based on performance and durability tests followed by in depth post-test analysis, and at the system level based on flow sheets and global energy efficiency calculations.

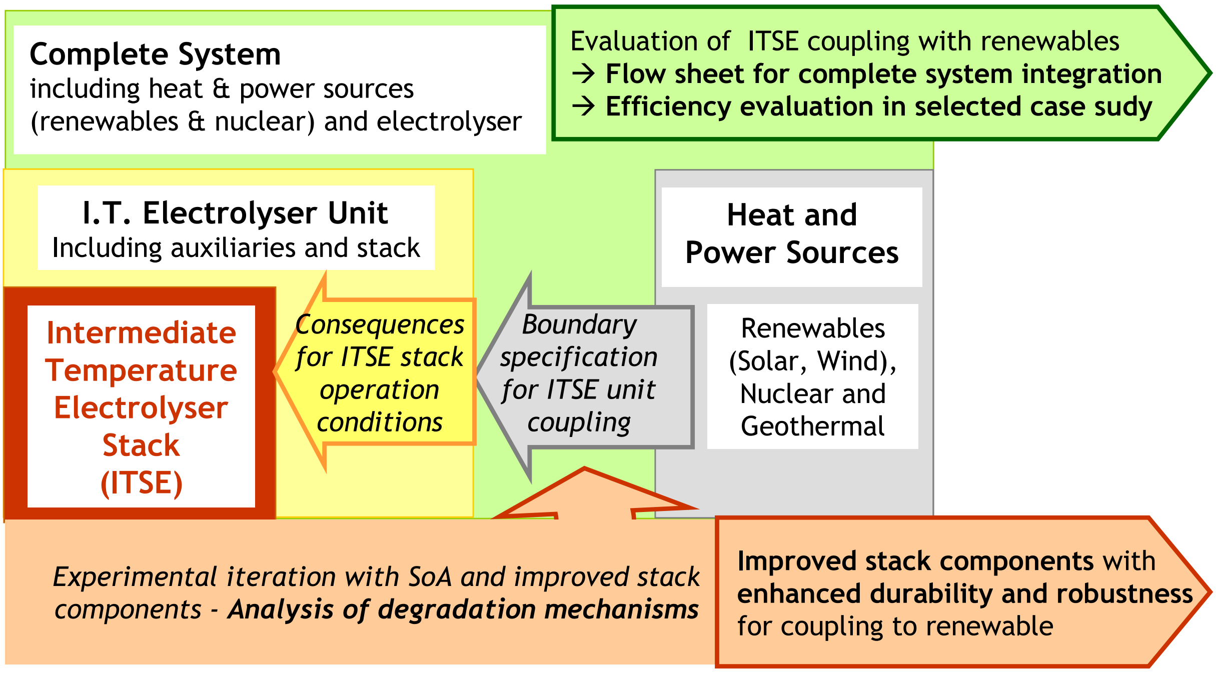

Figure 1: Concept of the ADEL project

Coupling of the Intermediate Temperature Steam Electrolyser unit (ITSE) with renewable or nuclear energy sources will in particular be studied from the point of view of the stack components to the complete system, flow sheets bridging the gap between the two scales.

1. Scientific Approach

The ADEL project targets the development of cost-competitive, high energy efficient and sustainable hydrogen production based on renewable energy sources. For such an ambitious target the project is built on a two scales parallel approach:

- At the stack level, the adaptation and development of cell, interconnect/coating and sealing components for ITSE operation conditions (T down to 600°C) aims at increasing the electrolyser durability.
- At the system level, the development of flow sheets to analyse and quantify the coupling between the electrolyser unit (based on stack data obtained at 600°C) and

renewable heat and power sources aims at identifying the most energy efficient solutions.

The quantitative assessment of the coupling relevance of the ITSE unit with renewable energy sources such as solar or wind or with nuclear and the preliminary dimensioning of a proof of concept technology demonstrator including an operating ITSE stack constitute the final outcomes of the project.

2. Rationale

High temperature electrolysis processes are considered in combination with various energy sources including wind, nuclear, solar, or geothermal energy, recommended by the European Hi2H2 project [3]. The integrated technology with energy sources previously cited is the focus of the ITSE development. The challenges are mainly focused on the following points [4]:

- recuperation, configuration, and optimization between the electrical cycle efficiency and the overall hydrogen production efficiency
- matching the operating conditions of each part of the plant to each other for optimal results, e.g. including storage and distribution of hydrogen

Indeed, depending on the temperature of the heat source and on the electrolyser operating temperature, the achievable proportion of energy that can be provided to the electrolyser as direct heat instead of electricity can vary significantly.

As illustrated in Figure 2, HTSE presents the advantage to accept direct heat (TΔS) as a complement to the electrical energy (ΔG) in the overall energy needed (ΔH) for hydrogen production. When the part TΔS of the overall energy needed (ΔH) is fully brought as direct heat, the process is said to be 100% allothermal whereas a 100% electrically driven process is said to be autothermal (with heat transfers provided by Joule effect).

An "allothermal ratio" has been proposed characterizing the proportion of TΔS which is provided by direct heat transfer ([5]): $\tau = T\Delta S_{\text{direct heat}} / T\Delta S_{(\text{total})}$ and leading to the following relationship:

$\Delta H = [\Delta G + (1-\tau)\, T\Delta S]_{\text{electrical}} + [\tau T\Delta S]_{\text{thermal}}$ with $\tau T\Delta S$ the direct heat contribution.

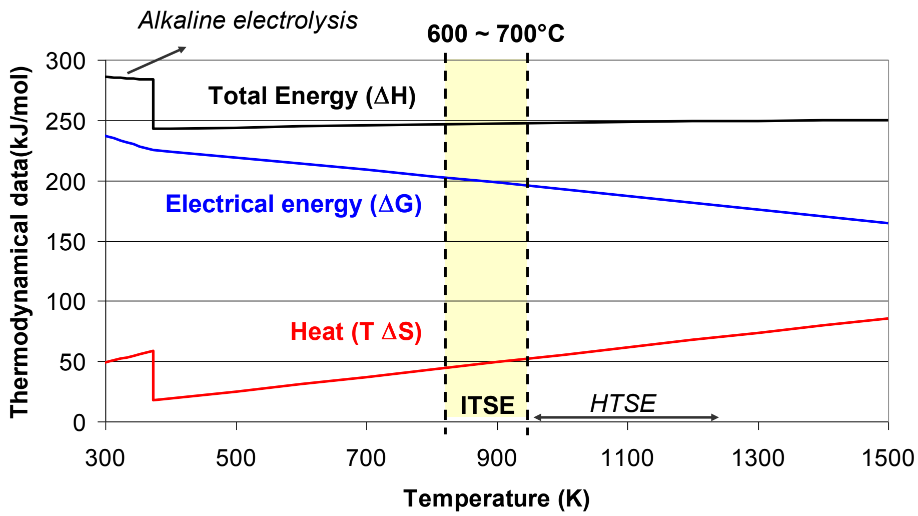

Figure 2: HTSE heat requests ΔH = ΔG + TΔS

Le Duigou et al have shown that the higher the allothermal ratio, the higher the energy efficiency [6]. Nevertheless, they have also shown that in the case of a HTSE unit operating between 810/850°C, an allothermal operation mode is possible only when coupled with a 950°C exit temperature source. The possible use of any present or future nuclear reactor generations (with exit temperatures around 200°C) or of renewable solar or geothermal heat sources (with exit temperatures about 230°C for geothermal and 800°C for solar) would suggest a "low temperature" coupling and an additional heating step leading to an auto-thermal operation mode.

Based on above considerations, the evaluation and development of intermediate temperature electrolyser cells is studied, aiming at stack component optimisation for increased durability. The project thereby benefits of its partners' experience from the intermediate temperature SOFC project SOFC600 [1] and High Temperature Steam Electrolysis (HTSE) activities like RelHy [2]. State-of-the-Art SOFC materials and individually evaluated components are integrated into short stacks and then tested under relevant operating conditions (high humidity, high current load, down to 600°C). After test, the components are analysed in order to establish the electrolysis-specific degradation mechanisms.

3. Experimental results

Two HTceramix/SOFCpower SOFC/SOEC short stack clusters (~300cm^2 total active cell area) were tested, stack one containing State of the Art ASC700 cells by SOFCpower, the second stack containing cells resulting from the EU FP6 project SOFC600. The stacks consist of an assembly of 6 Ni-YSZ supported cells with LSCF-CGO air electrodes (SoA) respectively with LSC-CGO air electrodes (SOFC600) and CGO barrier layers, interfaced with the proprietary SOFCONNEX™ gas diffusion layers and Crofer 22 APU metallic interconnectors. The latter are coated on the air side to reduce Cr evaporation. The cells differentiate slightly in the microstructural properties of the ceria barrier layer and the cathode support.

The test set-up in the SOEC mode is depicted in Figure 1. The stack is spring loaded at 0.6 kgcm^{-2} and placed inside a bell-furnace (Rohde, D). The steam is generated in an electrical evaporator (EBZ, D) supplied in water with a membrane pump (KNL, CH). The steam is then mixed with a H_2 flow (or H_2 and CO_2 in the co-electrolysis experiments) before entering the cathode compartment of the SOEC. All gas flows (air, N_2, H_2, CO_2) are controlled by Red-y mass flow controllers (MFC) (Vögtlin, CH). The pressure drop at the cathode and anode were measured with differential pressure transmitters (Jumo, CH). For the electrical circuit, a voltage source (24V, TDK) is connected in parallel to the stack to compensate its voltage and enable the active load to draw current. The latter was controlled with an electronic load (Agilent). The data (cell potentials, temperatures, pressures) were collected through a data acquisition system (Agilent) controlled by LabView.

Figure 3 depicts the performance of the SoA cell stack at temperatures at 600, 650 and 700°C. Performances starting from 650 and 700°C are judged reasonable, although not yet reaching the project targets, while the performance drop at 600°C is clearly marked and associated to slow electrode kinetics. To evaluate the degradation behaviour, this stack was operated for approx. 1'000 hrs at 650°C and 0.26 A/sqcm with 50% steam conversion. The individual cell degradation was on the average 3.7%/khr (voltage increase), ranging from 0.4 to 5.1%/khr for individual cells. The test was performed with anode flow of 50Nml/min/sqcm air and 4 Nml/min/cm2 (90% H_2O, 10%H_2) at the cathode

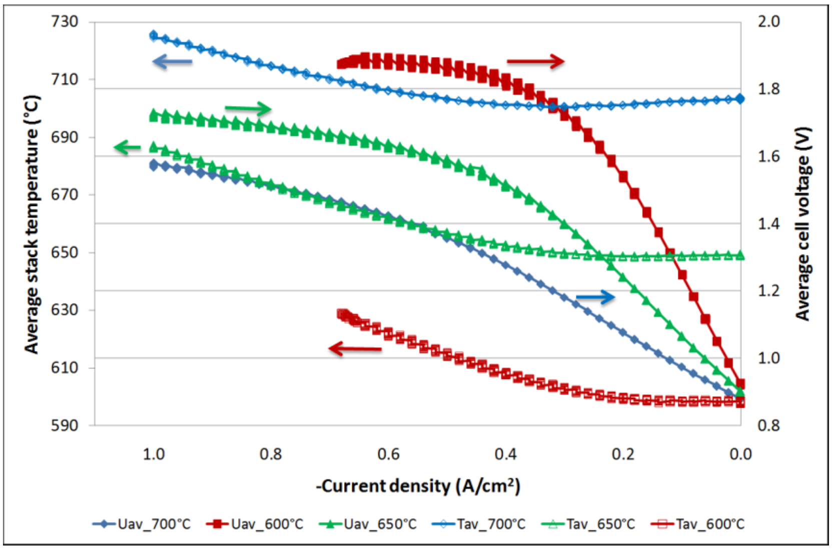

Figure 3: Initial V-i characterization in SOE mode at 600, 650 and 700°C

The initial performance comparison with the stack built with SOFC600 cells shows a clear improvement achieved by the cells developed for lower temperature application out of the SOFC600 project. At thermoneutral voltage and 700°C the improvement is close to 100% in similar conditions.

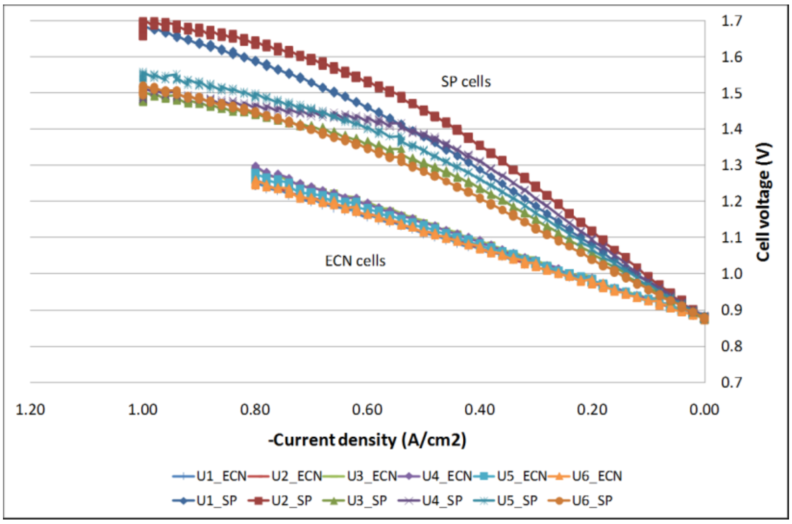

Figure 4: Initial performance comparison of SoA and SOFC600 cells in short stack

Short term degradation study shows a strong initial degradation that flattens with time, Table 1 shows the detailed figures, and figure 5 depicts the behaviour of the individual cells. This is the opposite phenomena to the one observed on stacks operated in SOFC

	Degradation rate/%/kh		
Cell	200-1324h	200-640h	750-1324h
1	9.8	21.7	3.24
2	0.9	-3.6	2
3	1.9	-2.2	1.9
4	2.1	-3.3	1.9
5	0.06	-2.2	0.9
6	4	3	4.8

mode.

Table 1: Overview of degradation of stack with SOFC600 cells.

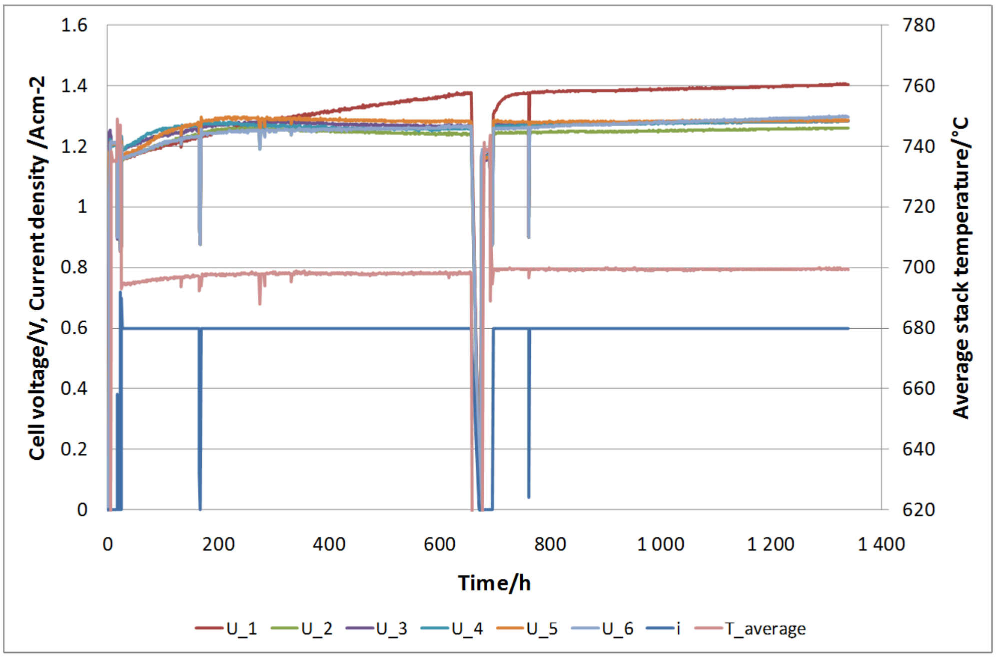

Figure 5: Steady state performance evolution of stack with SOFC600 cells in short stack

The degradation level is still too high with respect to the project and commercial targets. Post-operation analysis will be performed to identify the sources of degradation.

4. Summary of Workshop on SOE organized by ADEL

Within the ADEL project, an international Workshop was held in Sevilla [7]. 50 international experts exchanged and discussed their view on electrolysis in general and on SOE specifically. At the end of the 2 ½ days Workshop, the following conclusions were agreed upon between the experts:

For electrolysis in general:

- Hydrogen production from excess electricity is the key point
  - Intermittant/dispatchable operation is required
  - Grid balancing has an economic value
  - Intermode energy switch from electricity to mobility and/or heat reduces generally the carbon footprint
- Excess electricity-to-fuel by electrochemistry is of strong interest
- Electrolysis is a bridging technology and hydrogen is one energy vector towards low-carbon energy generation
  - Enabling more renewable and nuclear generation

For Solid Oxide Electrolysers:

- Electrolysis simulation and flow sheeting allow to orient materials search towards relevant objectives (T, p, i, durability)
- Simulation tools need to be validated against experimental performance

- Intermediate temperature stack operation (SOE@600°C) might not be required from a system point of view
- Pressurised SOE operation seems to be relevant from the system side
  - kinetically improved stack performance and reduced BoP costs
  - Does it affect degradation?

The participants qualified the information exchange as very fruitful. A second ADEL workshop will be organized in spring 2013, organized by CEA Grenoble.

5. Conclusion

Intermediate temperature steam electrolysis has a good potential to contribute to sustainable hydrogen production based on renewable energy. Reduced operating temperature opens the perspective of efficient thermal integration. Dedicated materials bring the stack performance close to the specified targets at 700°C, however the focal point currently resides in the identification of the specific degradation mechanism for cells in SOE mode.
From system point of view, operating under varying load conditions or also in grid dispatchable mode is critical and puts new constraints on materials, possibly also on stack design. An optimal performance and long-term stability point is researched. Further understanding of the underlying degradation mechanism is required.

6. Ackowledgements

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7. References

[1] http://www.SOFC600.eu/news/

[2] Florence Lefebvre-Joud, Marie Petitjean, Jan Peter Ouweltjes, Annabelle Brisse, Josef Schefold, Jacob R. Bowen, Sune D. Ebbesen, Ghislaine Ehora, Carlos Bernuy-Lopez and Jens Ulrik Nielsen, Proceedings of 9th Lucerne Fuel Cell Forum (Editor J. T. S. Irvine), 29th June to 2nd July 2010, Lucerne, Switzerland.

[3] Highly efficient, high temperature, hydrogen production by water electrolysis. Hi2H2 project, http://www.hi2h2.com/. June 2007.

[4] Herring JS, Lessing P, James E O B. Hydrogen production trhough high temperature electrolysis in a solid oxide cell. Second Information Exchange Meeting on Nuclear Production of Hydrogen, Argonne, Illinois. 2003.

[5] P. Lovera, A. Le Duigou, P. Carles, *Hydrogen Production Via Autothermal or Allothermal Processes*, presented at IHEC2007, 13-15 July 2007, Istanbul (Turkey).

[6] A. Le Duigou, P. Lovera, D. Haubensack, T. Gilardi, J. Paul-Joseph, G. Rodriguez, *Impact of High Temperature Steam Electrolysis Detailed Flow-Sheets on Massive Hydrogen Production Strategic Choices*, presented at IHEC 2007, ref. IHEC07-0106, Istanbul, 13-15 July 2007.

[7] http://www.adel-energy.eu/workshops.html.