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This document appeared in

Detlef Stolten, Thomas Grube (Eds.):
18th World Hydrogen Energy Conference 2010 - WHEC 2010
Parallel Sessions Book 3: Hydrogen Production Technologies - Part 2
Proceedings of the WHEC, May 16.-21. 2010, Essen
Schriften des Forschungszentrums Jülich / Energy & Environment, Vol. 78-3
Institute of Energy Research - Fuel Cells (IEF-3)
Forschungszentrum Jülich GmbH, Zentralbibliothek, Verlag, 2010
ISBN: 978-3-89336-653-8

Durability and Efficiency of High Temperature Steam Electrolysis as Studied in the RelHy Project

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1 Introduction

High Temperature Steam Electrolysis (HTSE) stands for a promising process for large-scale centralised hydrogen production and is also considered since the 1970s as an excellent perspective for efficient use of renewable energy sources [1,2]. Most of the water electrolysis technologies to date have used alkaline or acidic electrolyte systems with typical system efficiencies within the 55-75% range and current density typically around 0.3-0.4 A/cm^2 corresponding to 10-15 mgH$_2$cm^{-2} hr^{-1} [3-6].

Electrolysers based on solid-oxide cell technology offer the possibility of using heat generated from various sources in order to reduce the electric energy input and enhance the electrolysis efficiency. This had been demonstrated in the 80's by Dornier using tubular ceramic cells but for economical reasons no further development had been made. Nowadays, with environmental threatens, it is again considered as a promising carbon free hydrogen production mean and many R&D programs have recently being re-started worldwide [7,8]. However, HTE technology will be competitive regarding alkaline or PEM technologies only if an increase of the electrolyser life time can be obtained (> 3 years).

If hydrogen production rates as high as 100 mgH$_2$cm^{-2}hr^{-1} have recently been reported at 950°C with planar cells by [9,10] it has also been shown that the higher the performances the higher the degradation rates, especially in stack environment [11-12].

Main sources of degradation affecting the solid oxide electrolyser cells and stack lifetime are due to the high operating temperature (800 – 950°C). They have been studied and analysed in [13-18] and are principally:

- Delamination of O$_2$-electrode and bond layer on steam/O$_2$-electrode side,
- Poisoning of reaction sites due to Contaminants (Ni, Cr, Si, etc.),
- Loss of electrical/ionic conductivity of electrolyte,
- Mechanical break of the cell due to thermal cycling.

The RelHy project (www.RelHy.eu) launched in 2008 with 7 European partners from universities, technical research centres and industry, aims at reaching a satisfactory compromise between performance and durability and to demonstrate it at the electrolyser stack level. For such a purpose it is organised for identifying most detrimental degradation mechanisms at the scale of the single cell, the single repeating unit (SRU) and the short stack and for proposing solutions to overcome them.

2 Approach

Common test protocol has been established that comprises a first stage in the SOFC mode to reduce the hydrogen electrode and check the quality of the sample; a second stage in the SOEC mode at increasing steam partial pressures (from 50% to 90%) being repeated at 800°C, 850°C and 750°C; finally an endurance stage at 800°C and at current density below 1.5V but as close as possible to 1 $A.cm^{-2}$.

Common SRU to be distributed among the partners for testing have been developed (Figure 1). The main motivation for its design was to be as close as possible to the TOFC short stack design, to be easily mounted and instrumented by all the partners, to be easily simulated to assist data analysis. The 5-cell short stack (Figure 1) is based on the Alpha design by TOFC, it can accept planar cathode and electrolyte supported cells and has metallic interconnects.

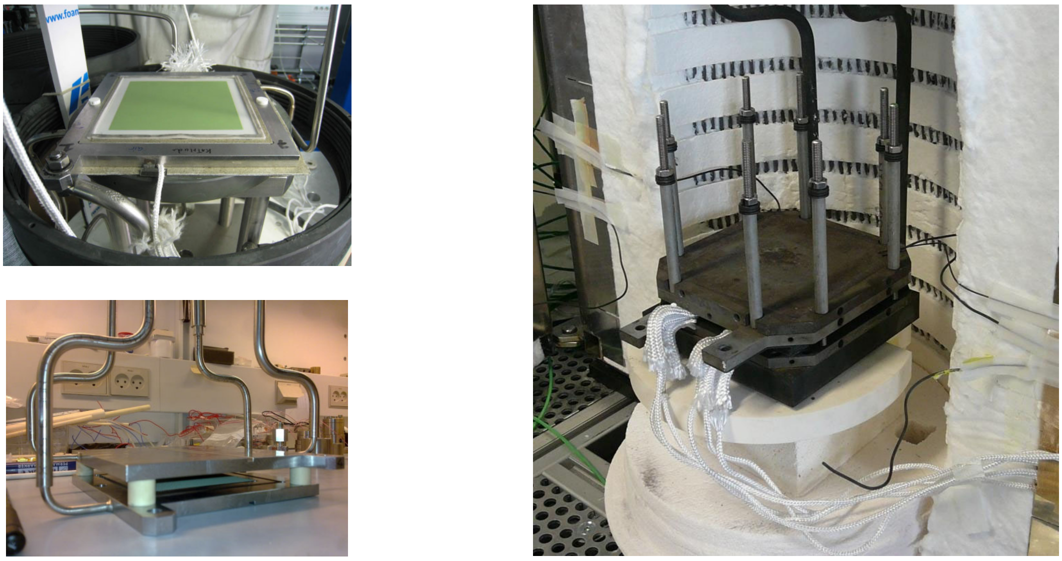

Figure 1: SRU upon mounting and 5-cell short stack ready for testing.

Tests are simulated by FE-CFD models integrating inputs from micro-modelling for data analysis, and extensive post-test characterisations is achieved to complement the analysis and identify the origin of degradation.

A first batch of materials chosen as representative of current state of the art has been tested in order to establish a reference basis within the project and to evaluate reproducibility within SRUs and between single cells, SRUs and short stacks. These reference materials include cathode supported cells Ni-YSZ/8YSZ/LSM-YSZ fabricated at Risoe-DTU and electrolyte supported cells Ni-CGO/3YSZ /LSCF fabricated at ECN. Interconnects are made with CROFER with LSM coating at the anode side and Ni based coating at the cathode side. For sealing reproducibility between testing laboratories, slurry options have been eliminated and the use of pre-sintered glass bars has been preferred.

3 Experimental Results

Performance and durability results of the reference materials have been obtained at the three levels considered with the two types of cells (Figure 2).

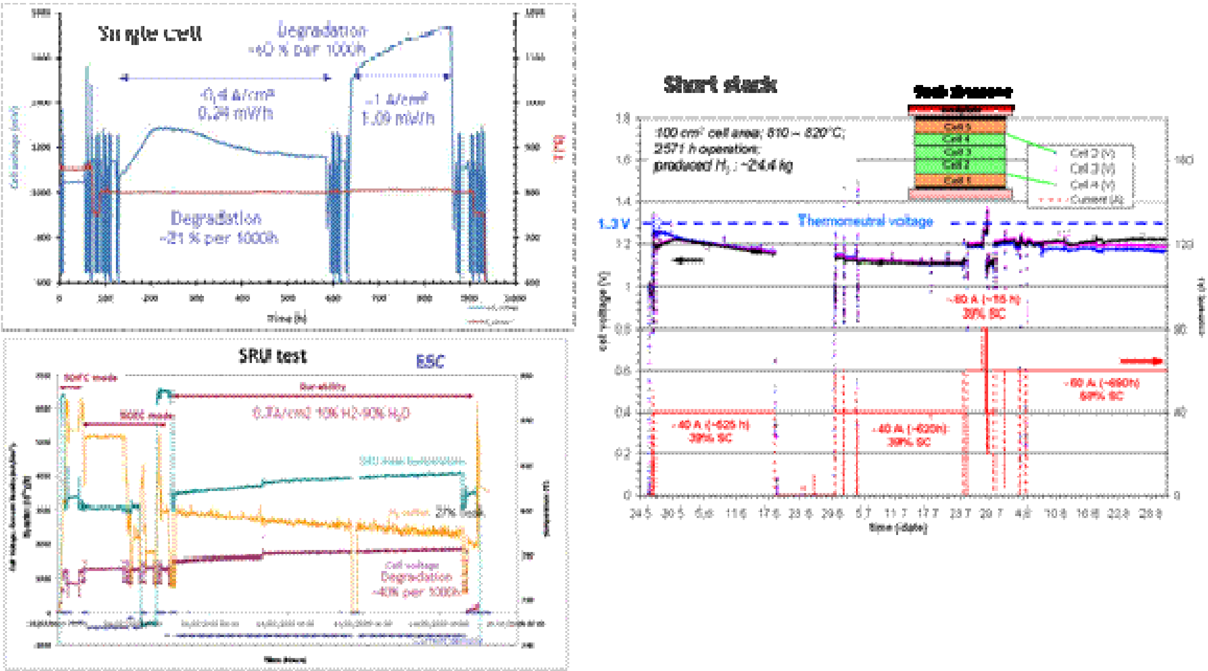

Figure 2: Duration tests at the single cell, the SRU and the short stack levels.

Scaling up from single electrolyser cell to short stack leads to different behaviour regarding performance and durability.

Regarding initial performances (Figure 3a), a tendency to degrade initial ASR when leaving grids for interconnect and single interconnects for short stack is observed; However, when comparing similar tests, a certain discrepancy is obtained at the single cell level as well as at the SRU level. Further tests have shown that satisfactory reproducibility between initial performances (at both levels) can be obtained when strictly controlling every operation parameter (Figure 3b); this being much harder in the SOEC operation mode than in the SOFC one.

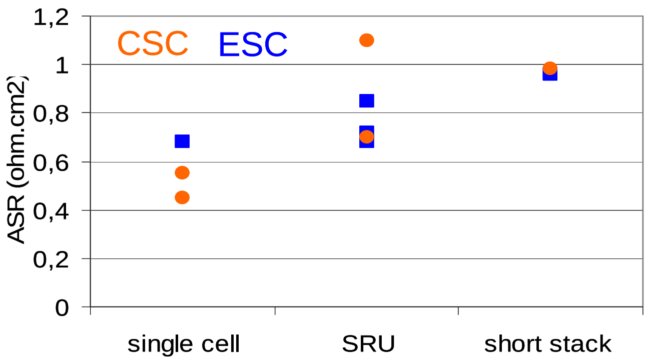

Figure 3a: ASR measured when starting SOEC operation at 800°C on both types of cells (ESC and CSC).

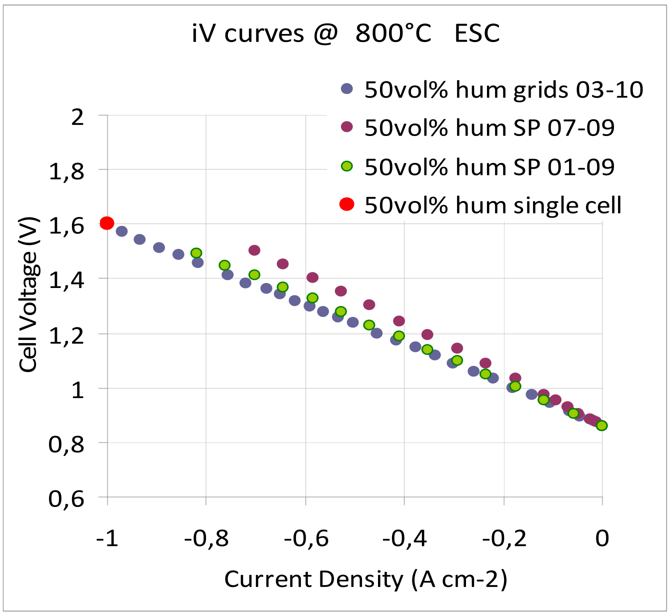

Figure 3b: Initial polarisation curves measured on an ESC at 800°C.
The same ASR ($0.7\Omega.cm^2$) can be obtained in a single cell test (red point) and in SRU tests (blue and green). Poor control of the electrical contact within the SRU leads to higher ASR (purple)

Regarding degradation, as an illustration, all results obtained on CSC are gathered on Figure 4. No clear tendency can be obtained since each result has its own "story". High degradation rates are obtained on single cells even at moderate current density and appear to be highly sensitive to test benches; high degradation rate has been obtained on a SRU at low current density that had poor initial ASR whereas more promising result has been obtained at 1A cm^{-2} with a SRU having a very good initial ASR. Low degradation rate is obtained at moderate current density at the stack level although initial ASR was not optimal. According to these results first order operation parameters on degradation are proposed to be:

- Electrical contacts within the single cell / SRU / stack since major differences have been obtained between the durability of similar cells with different initial ASR values;
- Steam generation and feeding since significant degradation events are always observed to be linked with steam feeding incident;
- Operation voltage that is found to be the most important parameter. Very low degradation rates (3% per 1000h) have been obtained at the stack level with cells operated below the thermo neutral voltage. On the contrary, at the single cell and the SRU level, operation above the thermo-neutral voltage have most of the time produced degradation rates higher than 10% / 1000h.

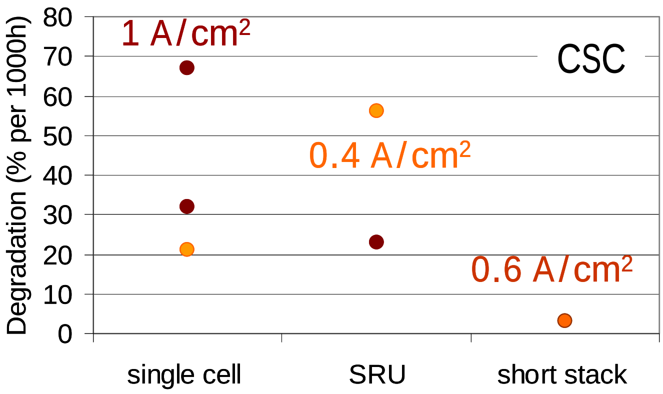

Figure 4: Degradation rates estimated over few hundred to 1 thousand hours of operation at 800°C on cathode supported cells.

4 Conclusion

Performance and durability results over few hundreds to thousand hours have been obtained on reference ESC and CSC following a specific testing protocol.

Good performance results could be obtained, as well as good durability at the stack level among 1000 hours.

Reproducibility between similar tests was found to be hard obtaining but main controlling parameters could be identified that are i) the electrical contacts with cells, ii) steam generation and feeding, iii) operation voltage.

Acknowledgments

The authors would like to mention and warmly acknowledge the essential participations of Marie Petitjean from CEA, Josef Schefold from EIFER, Bert Rietveld from ECN and Jens Ulrik Nielsen from TOFC to this work.

This work is part of the Relhy project funded by the European Commission in the 7th Framework Program (Grant Agreement 213009)

References

[1] M.M. Eisenstadt, K. E. Cox, Hydrogen production from solar energy, 1975 ; 17 :59-65

[2] Costogue E.N., Yasui R.K., Performance data for a terrestrial solar photovolatic/water electrolysis experiment, Solar Energy, 1977; 19 :205-10

[3] K. McHugh, Hydrogen production methods MPR-WP-0001, MPR Associates, Inc., Alexandria, Virginia, USA (2005)

[4] S.P.S. Badwal, S. Giddey and F.T. Ciacchi, Hydrogen and oxygen generation with polymer electrolyte membrane (PEM) based electrolytic technology, Ionics 12 (1) (2006), pp. 7–14

[5] P. Millet, F. Andolfatto and R. Durand, Design and performance of a solid polymer electrolyte water electrolyser, Int J Hydrogen Energy 21 (2) (1996), pp. 87–93

[6] M. Newborough, A report on electrolysers, future markets and the prospects for ITM Power Ltd's electrolyser technology

[7] Nuclear Hydrogen DOE USA

[8] European Fuel Cell and Hydrogen Joint Undertaking

[9] M. Mogensen, S. H. Jensen, A. Hauch, I. Chorkendorff, and T. Jacobsen. Lucerne (2006).

[10] S. H Jensen, P.H. Larsen, and M. Mogensen, Inter. J. Hydrogen Energy (2008).

[11] A. Brisse, J. Schefold, M. Zahid Inter. J. of Hydrogen Energy Vol 33, 5375, (2008)

[12] A. Brisse, A. Hauch, M. Mogensen, G. Schiller, U. Vogt, M. Zahid, Proceeding of the WHEC 2008 Brisbane Australia (2008).

[13] Manohar S. Sohal Degradation in Solid Oxide Cells During High Temperature Electrolysis, May 2009, Idaho National Laboratory, Idaho Falls, Idaho 83415, http://www.inl.gov, Prepared for the U.S. Department of Energy Office of Nuclear Energy Under DOE Idaho Operations Office Contract DE AC07-05ID14517

[14] A. Hauch, “Solid Oxide Electrolysis Cells – Performance and Durability,” Ph.D. Thesis,Technical University of Denmark, Risø National Laboratory, Roskilde, Denmark

[15] A. Hagen, R. Barfod, P. Hendriksen and Y. Liu, Effect of operational conditions on long-term stability of SOFCs , Proc.SOFC IX 1 (2005), pp. 503–513

[16] J.R. Mawdsley et al. Post-Test Evaluation of the Oxygen Electrode from a Solid Oxide Electrolysis Stack and Electrode Materials Development, AIChE Meeting, Salt Lake City, UT, November 5-9, 2007.

[17] J. R. Mawdsley, J. David Carter, et al. Post-test evaluation of oxygen electrodes from solid oxide electrolysis stacks. International Journal of Hydrogen Energy 34(9): 4198-4207.

[18] A. Hagen, R. Barfod, P. V. Hendriksen, Y. L. Liu and S. Ramousse, Degradation of Anode Supported SOFCs as a Function of Temperature and Current Load, J. Electrochemical Society,153(6), 2006, A1165-A1171