The aim of the DECODE project is to elucidate degradation mechanisms in PEFC with special focus on the influence of liquid water and in a second phase to modify components to achieve a significant improvement of PEFC durability. The focus of the project is the creation of new knowledge and understanding of the PEFC degradation processes, and in addition, the practical improvements of fuel cell performance and durability.

The project encompasses 11 partners with the necessary and important expertise to investigate and quantify degradation related phenomena in fuel cells and to derive strategies for improved durability. In particular, the project profits from the inputs of two car and truck manufacturers, component manufacturers, research institutes with their advanced testing infrastructure and universities with advanced modelling expertise. The project is structured into 7 work packages for the investigation of various components of PEFC and in the field of organization and dissemination (involvement of all partners). The work packages are shortly described at the end of the summary. DECODE focuses both on detailed component characterisation and also subsystem (short stack) testing and analysis. The project aims at deriving the maximum information from all testing and analytical work but also follows a pragmatic approach. If specific component information can only be derived from a specific model configuration it is accepted within DECODE that the simplified arrangement is used. On the other hand, components are tested in long-term operation in short stacks under realistic and technical conditions thereby leading to naturally aged components. These will be analysed with all available analytical tools within the framework of the project. The components of PEFC investigated in the project consist in the electrodes, membranes, diffusion media and bipolar stacks. In-situ and ex-situ analysis is performed leading ideally to preliminary life time predictions at the end of the project. A special strength of the project is the large modelling activity which is expected to significantly advance knowledge and understanding of the processes leading to degradation and also to deliver the tools to describe ageing and performance degradation.

Progress of work since start of the project

At the start of the project it was of imminent importance to specify the requirements, materials and components as well as test procedures used and applied within DECODE. Also a definition of test cells and stacks for all activities was of paramount importance in order to ascertain a coherent activity and comparable results. In the discussions, which are organized within WP2, it was decided to follow a pragmatic approach to enable the comparability of results but also to ascertain the rapid establishment of testing capability within DECODE. Therefore the specifications were performed specifically for each work package taking into accounts the different requirements and degradation mechanisms effective in the various components of PEFC. The work of this WP is now finalized and summarized in the deliverable reports “D2.1: Report which materials and components are selected, description of the test cell for standard test” and “D2.2: Report about test conditions for standard tests incl. operation modes”. The specifications started from input given by the automotive industry about operation conditions relevant for automotive applications. Based on these conditions, test conditions were defined for work package 3-5 for standard tests that should at the same time especially enhance the degradation of the component investigated in that WP (membrane, reaction layer, gas diffusion layer and cell) but also ensure a comparability of results between the different work packages. The test procedures for continuous operation and dynamic operation (load cycling) are based on the results of the FCTES^{QA} project. Also the project test cells and stacks were defined. For single cell measurements every partner uses its own single cell, mostly specialized for certain measurements. For stack measurements DANA provides the material in form of short stacks. A membrane by Solvay-Solexis and a GDL by

SGL were defined as the single cell standard material. As catalyst coated membranes (CCM) were not available for tests as originally expected, tests with CEA home made CCM and commercial CCM by Solvicore had to be performed before the project test material could be defined. These tests are also terminated now.

According to the project plan the specification phase is be terminated at this point, but it is clear to all partners that modifications to these specifications may be necessary in the future with knowledge gain and shifting focus from understanding to improvement. Therefore updates of the specifications will be necessary and will be reported in future.

For all components state-of-the-art reviews (reports) are being prepared to have a solid knowledge base from literature to derive the strategies for the improvement phase where strategies to incorporate durability gains are derived. Due to the delayed start of the project but also by the size and extension of the available literature on this topic with many ramifications the reports have been delayed significantly. Most or the reports are available in the final state and no significant impact of the delayed on the project success is expected from the delayed deliveries. The reports on the state-of-the-art will be made public by peer reviewed publications of the project in a suitable journal. For this peer reviewed publications the content of the deliverables combined D3.1 and D3.2 related to the degradation of CCM, D4.1 (degradation of GDL) and D5.1 (degradation of bipolar plates) will be used as base.

The first focus of the membrane and electrode activities has been the definition of the CCM material based and the testing procedures to ascertain a good comparability and successful testing in all laboratories. This work which has been mainly preformed in collaboration between CEA and Solexis yielded first ageing results of differently prepared CCM but – more important- defined the reproducible material basis for further investigation at all DECODE laboratories involved in membrane and electrode investigations. Now commercial MEAs and MEAs prepared by CEA are available for comparative in-situ tests and laboratory scale CCM formulation is also available for ex-situ characterizations. The study of the degradation of the MEA at different stress factors is still running. Solvay Solexis performed the synthesis and extensive characterisation on the AQUIVION™ membrane used within the project including proton conductivity, water uptake, mechanical stress and hydrogen crossover measurements. In addition, performance data of CCMs and chemical and physical characterisations of the necessary ionomer dispersion are available. The membrane was also preliminary characterised by CEA with SANS (small angle neutron scattering). The necessary detailed analysis to provide indications for further developments to Solvay-Solexis (related to deliverable D3.3) should be possible. Samples from operated single cell (electrodes, membrane and CCM) from different positions inside the cell were delivered from CEA to DLR for further ex-situ investigations.

A special focus of the DECODE project is on advanced modelling of porous media and the experimental characterization of gas diffusion layers. On major modelling activity, discussed in this paragraph, is the use of the relatively novel Lattice-Boltzmann technique for the description of porous media and complex fluid systems. In the Lattice-Boltzmann models the fluid consisting of fictive particles, and such particles perform consecutive propagation and collision processes over a discrete lattice mesh. Due to its particulate nature and local dynamics, LBM has several advantages over other conventional CFD methods, especially in dealing with complex boundaries, incorporating of microscopic interactions, and parallelization of the algorithm. An adaptation of lattice-Boltzmann tool waLBerla for an efficient parallelisation of the free-surface application was performed. Key assignment for this task is a local management of gas volume changes, and merge of independent gas regions. In

addition also new algorithms for describing diffusion in porous media were developed. As a basis for the computational domains of the molecular dynamics of the MPL and the lattice Boltzmann model of the substrate, Opel provided a 3D synchrotron radiography data set of SGL GDM and handed it out to the partners who will convert it into computational domains for their respective models. To make the domain generation more convenient, Opel also provided a small set of image data carrying all relevant features of the original GDM image. Furthermore, Monte Carlo simulations of porous media are an important topic of DECODE based on structural models of the porous media. Three different type structures: fiber-, paper- and spaghetti- structures were generated and investigated. The distribution of PTFE was investigated on model surfaces and the models were verified through plausibility considerations of liquid water interactions. In addition, a pore network modelling tool (CEA) and a tool adapted from molecular dynamic modelling (UER) were developed and used for the simulation of transport processes.
A 3D pore network modelling tool (PNM) has been developed to simulate two-phase flows inside the GDL backing. The inputs are the pore size, the throat link size and the wettability distributions inside this porous media. The outputs are the two-phase pattern inside the GDL as well as its effective properties such as capillary pressure, relative permeabilities, gas diffusion as a function of liquid saturation. As the main degradation mechanism for the GDL is hydrophobicity loss, the degradation scenario studied by PNM is the increase of the number of hydrophilic pores over time. Results clearly show the existence of a threshold below which degradation has not a big effect on the GDL fluidic's behaviour; above this threshold, hydrophobicity loss induces a strong decrease of effective properties, especially gas diffusion, that will reduce gas access to the active layer and lead to flooding of the PEMFC. The different models to describe the gas transport in the GDL were reported in the deliverable 4.3.

GDL experimental characterization was performed by various methods like x-ray photoelectron spectroscopy, infra red spectroscopy, water porosimetry (in development) of artificially and naturally aged GDLs. Furthermore, neutron and synchrotron imaging and tomography results of fuel cells with relevance to GDL behaviour are generated in other projects, but can be used for DECODE. The methodological basis for investigation of porous media is exceptional in this project. It is expected that the experimental validation of the models will be achieved and that novel scientific results will be generated. This is especially important as the advanced characterisation of the diffusion media is lacking behind compared to the other fuel cell components. Artificial aged GDL materials were prepared by chemical and radiation treatment. The degradation of these artificial aged GDL was investigated by ex-situ methods. In addition natural aged GDLs from the short stack experiments in WP5 are available for single cell tests.

For the investigation of bipolar stacks DANA has provided 5 cell short stacks for Round Robin tests and for durability test. The first ageing results have been generated and are being analysed. It is also planned to use the GDL in the stack for investigating naturally aged GDL (link to WP4) with the methods described above. Within the project, an existing bipolar plate design "FRIEDA" from DANA Victor Reinz was selected for the test stack setup. The tested materials are uncoated SS316L, SS316L with 100nm electroplated gold coating, uncoated SS905L, and milled graphite composite. It is planned to iterate the different materials at different partners for testing under same conditions. The conditions of the durability run were defined within the DECODE Project with input from DLR, Opel and DANA. The first 1000 h durability test have been performed and the materials have been analysed. The tests are still in progress at the partners DLR, CEA, VOLVO, ZSW and DANA. 18 short stacks have be tested by the partners. The first results are reported in the deliverable D5.2.

Up to now ten stacks have been tested and fully analyzed. Twelve stacks finished the durability run and the post mortem analysis of the components is ongoing. One stacks to compare the test benches from each partner (round robin test) are still in operation. As well as the ongoing analysis of a single cell stack which was analysed under the lead of ZSW in the CONRAD (Cold Neutron Radiography) setup in Berlin.

The characterization and investigation of bipolar plates in durability measurements has started already. Presently, the components of the operated stacks are analysed with physical methods with various physical methods in order to identify the alteration induced by degradation processes. Contact resistances, water analysis and ex-situ surface analysis have been performed and the determination of trends and relationships are still ongoing.

Several publications and presentations concerning DECODE have already been realized. These are summarized below. DECODE was present in the "International Workshop on Accelerated Testing in Fuel Cells" with contributions of different partners (CEA, DLR, DANA, JRC etc.). The coordinator was in the scientific committee of the work shop and the DECODE logo was on all announcements.
DECODE was presented at:
"International work shop on Accelerated Testing in Fuel cells" in Ulm, Germany in October 2008.
"EUCAR-Workshop" in Brussels in September 2008
"European Fuel Cell and Hydrogen week" in October 2008
"EUCAR Conference & Reception at Autoworld" in Brussels on November 26th, 2008.
In Journal HZwei 01 2009, on page 16-17.
Further publications are

- M. Holber, A. Carlsson, P. Johansson, L. Jörissen, P. Jacobsson; *Raman investigation of degradation and aging effects in fuel cell membranes*; ECS 216, Vienna, Austria, Oct 4-9, 2009, in press.
- S. Donath, C. Feichtinger, T. Pohl, J. Götz, U. Rüde, R. Biswas (Eds.); *A parallel free surface lattice Boltzmann method for large-scale applications*; Parallel CFD 2009, 21st International Conference on Parallel Computational Fluid Dynamics, May 18-22, 2009, Moffett Field, CA, USA, pp. 198-202. (21st International Conference on Parallel Computational Fluid Dynamics, Moffett Field, CA, USA, 18.05.2009-22.05.2009), pp. 198-202.
- S. Donath, C. Feichtinger, T. Pohl, J. Götz, U. Rüde, H. Sips, D. Epema, H.-X. Lin (Eds.); *Localized parallel algorithm for bubble coalescence in free surface lattice-Boltzmann method*; In: Sips, H.; Epema, D.; Lin, H.-X. (Hrsg.): Euro-Par 2009 (Euro-Par 2009, Delft, 25. - 28.08.2009) . Berlin Heidelberg, Springer, 2009, p. 735-746 (Lecture Notes in Computer Science, Vol. 5704)
- F. Deserno, K. Iglberger, C. Feichtinger, J. Götz, S. Donath, C. Mihoubi, U. Rüde; *A geometry tool for waLBerla;* Technical report 09-12, Friedrich-Alexander University of Erlangen-Nürnberg, 2009.
- R. A. Neher, K. Mecke, H. Wagner; *Topological estimation of percolation thesholds;* Journal of Statistical Mechanics: Theory and Experiment P01011, 2008.
- S. Münster, B. Fabry, A. P. Sheppard, L. M. Jawerth, D. A. Vader, D. A. Weitz, W. Mickel,
  K. Mecke, G. E. Schröder-Turk; *Robust pore size analysis of filamentous networks from three-dimensional confocal microscopy;* Biophysical Journal 95, 6072-6080, 2008.
- H. Hansen-Goos, K. Mecke; *Fundamental measure theory for inhomogeneous fluids of non-spherical hard particles;* Physical Review Letters 102, 018302, 2009.

- K.A. Friedrich, M. Schulze, A. Haug; *EU - Project DECODE - Understanding of Degradation Mechanisms to Improve Components and Design of PEFC;* Hydrogen & Fuel Cells 2009 - International Conference, Vancouver, Canada, 31.05.09-03.06.09
- M. Schulze, A. Haug, E. Gülzow, K.A. Friedrich; *Investigation of Local Degradation Effects*
  Fuel Cell Seminar 2009, Palm Springs; ECST in press

In October 2009 a workshop together with the EU-project AUTOBRANE were performed.

In general, the project has been successfully started and all work packages are active in the tasks planned. There are significant delays in some tasks, in particular in the preparation of the state-of-art-reports. Delays are also evident in the investigation of electrodes and membranes in CCMs. However, the overall goals are not endangered and it is expected that the delay can be completely recovered in 2009.

DECODE is organised within work packages, which are:

Work package 1: Project management and coordination
Organization and management of the project

Work package 2: Requirements and Specifications
Definition of operating conditions, materials and testing procedures. Common understanding of the test capabilities and resources.

Work package 3: Investigation of Membrane and Electrodes degradation
Understanding of PEFC membrane electrodes assembly degradation under steady-state and transient operating conditions representative of automotive applications.

Work package 4: GDL Degradation Characterization and Assessment
Understanding of fundamental degradation mechanisms relevant to the porous media in PEFC. Models and thinking tools for the description of the effects of liquid water in porous media are developed in order to describe microscopic and nanoscopic effects.

Work package 5: Investigation of Degradation of Bipolar Plates and Seals
Understanding the fundamental degradation mechanisms of metallic bipolar plates with/without appropriate coatings as well as of bipolar plates manufactured of composite materials.

Work package 6: Improved durability
In the work package 6 the results and ideas from the work packages 3, 4 and 5 will be used to improve durability of fuel cells. Starts in month 21st.

Work package 7: Recommendation and Dissemination
The objective of the work package 7 is to disseminate the results for the scientific work.