Reducing Fuel Cell Degradation in Micro Combined Heat and Power Systems

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Abstract: Fuel cells in domestic micro combined heat and power (μ CHP) systems are subject to several degradation phenomena that can be counteracted with appropriate control strategies. Cost is an important factor, and additional equipment must be minimised. Controllers are proposed to counteract stack drying, flooding, poisoning by CO, and to estimate anodic hydrogen excess ratio to prevent hydrogen starvation. Tests on an emulator in LabVIEW indicate that the controllers are able to perform their function, and will be tested on a real system in 2015.

Keywords:fuel cell; degradation; flooding; drying; CO poisoning; hydrogen starvation; control system synthesis

1. INTRODUCTION

Micro Combined Heat and Power (μ CHP) systems based on fuel cells are designed to replace the common domestic natural-gas heaters used in many countries worldwide. Their advantage over more common burners is that, in addition to heat, they also produce electric power, which is usually priced much higher than natural gas on an energy basis.

Fuel cells are devices that convert the chemical energy of some fuel, typically hydrogen, directly in electrical energy, i.e. without combustion. They are well suited to be employed in μ CHP systems because of their high efficiency, scalability and silent operation.

A significant disadvantage of fuel-cell μCHP is the cost of fuel cells and their limited lifetime, which has hampered market adoption in the EU. Japan has instead seen a wide adoption of domestic μCHP , with about 100 000 units sold by 2014, which is mainly thanks to government subsidies. SAPPHIRE 1 is a research project funded by the EU to increase the lifetime μCHP fuel cells by means of advanced control.

We here consider a commercial μ CHP system produced by Dantherm Power A/S². Dantherm Power is a Danish company specialised in the integration of low-temperature PEM fuel cell solutions for telecommunications power back-up, μ CHP, fuel cell systems for small industrial vehicles and system engineering services. The low-temperature proton exchange membrane (LT-PEM) fuel cell stacks of the μ CHP are typically sized to deliver about 1–2 kW electrical power. The hydrogen fed to fuel cells is produced from natural gas in a reformer. The system produces excess heat that is made available for the consumers' direct heating needs by hot water production. The focus of this work is on the fuel-cell stack, so the internals of the fuel processor and its control structure are not elaborated in detail.

Fuel cells are subject to degradation of their performance over time, and limitations in their lifetime increase the cost of ownership of a μ CHP system. The lifetime of such a fuel-cell system can be increased with appropriate control action. In this paper, we consider the current control layout of a present-generation μ CHP system and propose techniques to reduce degradation and, in perspective, to integrate prognostic techniques.

2. BACKGROUND

2.1 The μCHP system

Layout and Description This section recapitulates a more detailed description of the μ CHP system as described by Menard (2013).

The μ CHP system by Dantherm Power is sketched in figure 1. Water from a reservoir tank is vaporised and mixed with natural gas before entering a fuel processor, which consists of a reformer and several purification units; these produce hydrogen and CO₂ according to the overall reaction:

$$CH_4 + 2H_2O \longrightarrow CO_2 + 4H_2$$
 (1)

where for simplicity natural gas was assumed to be pure methane.

Water vapour is usually present in excess in order to minimise the formation of CO, which is a catalyst poison for the fuel cell; CO and water vapour can react through what is called the water-gas shift reaction:

$$\rm CO + H_2O \longleftrightarrow \rm CO_2 + H_2$$
 (2)

The produced gas ("reformate") is fed to the stack's anodic side after being mixed with a small quantity of air ("air bleed"), with the objective of eliminating the last CO that may have remained in the reformate flow by oxidising it to $\rm CO_2$ on the anode catalyst. Low-temperature fuel cells can

 $^{^1}$ See sapphire-project.eu.

 $^{^2}$ See dantherm-power.com.



Fig. 1. A flow diagram of the μCHP system.

tolerate only minimal levels of CO concentration, typically a few ppm.

The effluent from the stack still contains some hydrogen, which is then reacted in a burner to heat the vaporiser and provide therefore the temperatures necessary to the functioning of the fuel processor. The burner effluent is then passed through the co-generator (heat exchanger) to recover its heat, and finally (not shown in figure 1) condensed water is recovered and gathered back in the water tank.

The cathodic side of the stack is fed by environment air, after it has been heated and humidified by the cathodic effluent; the humidifier is a passive unit and cannot be controlled. The effluent proceeds to the co-generator to recover the last part of its heat.

Heat from the stack is removed by a cooling loop that connects directly stack and co-generator. The co-generator itself receives cold water from the return pipe of the household heating system, and heats it with waste heat from the stack (through cooling water and cathodic effluent) and from the exhausts of the fuel processor's burner.

Electrically, the stack is connected to an inverter unit that converts the produced DC power into 1-phase, 220 V AC power, which is fed to the electricity grid.

Regulatory Layer For industry actors such as Dantherm Power it is a priority to reduce the number of sensors and actuators in the μ CHP system in order to reduce its cost. Currently, the system allows to measure several temperatures, gas and liquid flows through every pump, and stack voltage. The actuators are the gas and liquid pumps and the power control system, which allows to set the stack current directly. Very few sensors are present on the stack itself, and the SAPPHIRE project has a cost target of less than 100 /kW for additional equipment: the possibilities for modifications of the process are therefore very limited.

The present control system consists of a regulatory layer that sets gas and liquid flows according to some preset values, which all ultimately depend on the power output.

The system is designed to provide hot water to a household, and to run at nominal power for the winter months; in summer, the system is switched off as the household cannot absorb all the heat it produces. In this perspective, electric power is a useful by-product of heat generation; it is assumed that the μ CHP unit is always connected to the grid, which can always accept its power production.

2.2 Degradation Pathways of Fuel Cells

There are multiple degradation mechanisms in fuel cells, each with its specific conditions for their occurrence, but only a few ways to detect them during operation: most often gradual voltage loss, sometimes complete and sudden failure without warning. Bezmalinović and Barbir (2014) have given a detailed account of degradation phenomena in low-temperature PEM fuel cells, which will be recapitulated here in its relevant aspects for this paper.

Degradation phenomena can be reversible or irreversible; by definition, only irreversible degradation should be considered to determine the residual useful life of the stack. However, even reversible degradation can lead to permanent damage if left untreated for long periods of time, typically by enhancing stress factors of irreversible degradation patterns.

Reversible degradation's immediate effect is that it disrupts the operation of the stack, and this is the main reason why it is important to devise a controller able to promptly counteract its effect. This paper considers the following three reversible degradation patterns for lowtemperature PEM fuel cells:

- **Drying:** dry conditions increase ohmic resistance in the membrane and reduce catalytic activity. An appropriate corrective action is lowering the stack temperature, so that the relative humidity will increase.
- Flooding: too humid conditions can result in excessive production of liquid water on the cathode, which can obstruct the access of oxygen to the reaction sites. To correct the condition, temperature can be increased to promote evaporation.
- Catalyst poisoning: unconverted carbon monoxide from the reformer can significantly reduce the activity of the fuel cell's anodic catalyst. Corrective action is to increase the air bleed to oxidise CO to CO₂.

It should be noted that all these phenomena result in a reduction of measured stack voltage, and cannot be distinguished without further analysis. In particular, the appropriate corrective action for both drying and flooding is to restore the correct humidity level: this can be done by respectively reducing or increasing the cell temperature, which is usually controlled in an inner cascade loop by manipulating the flow of coolant. The importance of distinguishing the two phenomena is then evident, since the solution to one worsens the other. The normal humidity level in a LT-PEM is just above 100 %, with some droplets forming on the cathode.

Electrochemical Impedance Spectroscopy (EIS) is a proven and effective way to correctly diagnose which phenomenon is occurring (Le Canut et al., 2006), but it requires expensive and cumbersome equipment to perform the EIS. For commercial μ CHP systems, it is necessary to develop more cost-effective estimators of temporary degradation phenomena.

Yousfi Steiner et al. (2011), for example, used neural networks to model voltage and pressure drop as a function of readily measurable parameters; if measured voltage was found to be low, a fault condition was identified; with a high pressure drop, it was deemed to be flooding, otherwise drying. Whereas this method does not require EIS, it does assume training of a neural network for a specific stack, which is a resource-intensive task.

In LT-PEM fuel cells there are several irreversible degradation phenomena, but for the scope of this paper we will consider only one, **hydrogen starvation**. This occurs if not enough hydrogen is provided to the cells to sustain current, in which case the cell potential rapidly falls. When cells are arranged in a stack, which is electrically connected in series, one cell subject to hydrogen starvation may still be forced to pass current by other cells in the stack. In the absence of hydrogen, the carbon catalyst support is consumed as fuel in reverse electrolysis, which results in a very rapid and permanent degradation of the anodic catalyst. Therefore, hydrogen starvation is a condition that must be avoided at all times, by monitoring the hydrogen excess ratio λ .

3. PROPOSED CONTROLLERS

In this section, several control strategies are proposed to detect, discern and counteract degradation phenomena.

3.1 Drying

It has previously been observed in a study of single cells (Barbir et al., 2005) that drying results in higher ohmic internal resistance of fuel cells. This effect can be measured with appropriate electronic equipment, but this would be expensive and require resized electronics for different sizes of μ CHP systems.

In the same graphs reported by Barbir et al., another phenomenon is however clearly visible: as humidity reaches below 100% in the cell's cathode, resistance to air flow drops, and the disappearance of droplets makes the pressure drop very steady (air flow is always in laminar regime).

A differential pressure measurement between cathode inlet and outlet can be installed to continuously measure flow resistance, or pressure noise level, or both: drying will then be detected by a low measured value or noise in pressure drop. Such measurements are cheap, reliable and can be applied to a wide range of μ CHP systems.

When drying is detected, a natural course of action is to reduce the stack temperature by increasing the cooling loop flow. A PI controller may be tuned for each specific μ CHP system to counteract cell drying.

Since the air flow takes the path of least resistance, the effect of one dry cell in a large stack will be magnified as more air will pass through that cell; this method should therefore be more appropriate to spot drying at an early stage, when it only involves few cells.

3.2 Flooding

Barbir et al. (2005) also considered the detection of flooding in a single cell, suggesting to monitor pressure drop over the cathode, and correlate increases in flow resistance with flooding.

There are however several shortcomings to this approach, in particular when applied to a stack:

- Air flow takes the path of least resistance: flooded cells will receive less air, and their pressure drop will be reduced;
- The increase in pressure drop indicated by Barbir et al. is not large, and will be difficult to discern from random variations in case only few cells in a stack are flooded.

Pressure drop is therefore not considered an appropriate diagnostic variable for flooding in stacks.

However, cell voltage presents wide oscillations when the cell is flooded, which do not occur in normal operation or other temporary degradation. Stack voltage is the sum of the voltages of all cells, which are connected in series. Cell voltages are essentially independent from each other, so that if each cell is flooded and exhibits a voltage standard deviation σ_V , the standard deviation of the total stack voltage will be $\sqrt{n} \sigma_V$, e.g. just about 7 times larger than the standard deviation of a single cell in the case of a stack with n = 50 cells. This means that the noise from one or few flooded cells will be much easier to discern early and therefore to compensate for.

Just like for drying, it is possible to tune a PI controller to adjust the stack temperature when flooding occurs; indeed it may very well be the *same* PI controller, with the only difference that temperature will need to increase to promote evaporation.

3.3 CO Poisoning

The equilibrium reaction 2, the water-gas shift, implies that the gas leaving the fuel processor will contain a certain amount of CO. Fuel processors are therefore set up with several sub-units that gradually reduce the CO content down to ppm-level concentrations, but it is still possible for some amounts of CO to reach the fuel cells. In particular, in case of dynamic load, temperature transients in the units can result in surges of CO that can have a significant impact on the operation of fuel cells.

CO poisons the catalyst by occupying the reaction sites that hydrogen uses to dissociate into protons and electrons; CO's adsorption characteristics make it occupy almost all catalytic sites, given enough time. When a large fraction of these sites are covered by CO, and only a small fraction is available to hydrogen, voltage losses in the stack become noticeable.

A complete set of dynamic equations describing the CO poisoning process is given for example by Kadyk et al. (2011). When simulating the transients in CO poisoning, it is noticeable how a small concentration of CO (1-10 ppm) of the kind that is normally expected from a fuel processor can take hours to reach steady state.

To compensate for CO poisoning, it is industry practice to add an *air bleed* flow to the anode, i.e. a small percentage of air to the fuel flow (regulated well below the explosive threshold). The oxygen in the air oxidates rapidly CO to CO_2 , which then leaves the catalyst free for hydrogen to dissociate on. The oxidation of CO by O_2 is generally a very quick process (a few seconds), and if CO poisoning is present an upwards jolt in voltage is usually well visible.

At the same time, too much air bleed is undesirable because it also consumes hydrogen fuel; the chemical reaction with hydrogen on the catalyst sites also generates a large amount of heat, which over time leads to degradation of the catalyst.

Using air bleed against CO poisoning is therefore a process with highly asymmetrical dynamics: voltage increases rapidly with air bleed, but decreases much more slowly.

To counteract CO poisoning, the following procedure is proposed:

- (1) The stack voltage and its noise are measured over a standard period of time, typically 30 seconds;
- (2) An increase in air bleed percentage is performed, typically 0.2 %;
- (3) The voltage and its noise are again measured after the increase in air bleed;
- (4) If the voltage has increased more than what can be explained as random noise, the bleed increase is assumed to have removed CO; the algorithm is immediately repeated from point 1.
- (5) If no voltage increase is measured, or if the increase is within the noise level, air bleed is reduced by 150% of the original increase (therefore, typically 0.3%).
- (6) The algorithm is then left dormant for a time sufficient for CO poisoning transients to occur, typically 6 hours, and then restarts from the first step.

Step 5 is necessary to allow the controller to reduce air bleed as well as to increase it. Due to the discrete nature of this procedure, air bleed will not settle to a steady state, but oscillate periodically around the threshold where CO poisoning occurs.

The presented method is able to compensate for variable CO poisoning without a costly CO measurement, using only the readily available stack voltage measurement. A weakness of the controller lies in the long hibernation times necessary to wait for CO adsorption transients to complete (step 6): if a large surge in CO were to occur, it would completely block the catalyst before the air bleed had a chance to compensate for it, since CO poisoning transients are significantly faster at higher concentrations.

3.4 Hydrogen Starvation

Hydrogen starvation occurs when a cell draws more current than what is sustainable by the available hydrogen. When hydrogen depletes and current is still applied, the catalyst in the depleted cell will undergo rapid degradation of the catalyst by reverse electrolysis.

To avoid starvation, hydrogen is fed to the cell stack with an excess ratio λ between 1 and 2, the exact value depending on the specific system. The presence of other products from reforming (water vapour and CO_2) makes it necessary to extract these gases from the stack, along with the excess hydrogen. Since the flow of outlet hydrogen still has a significant energy content, it is used in the burner of the fuel processor to provide the heat necessary to run the reformer.

Using a chemical sensor to determine the quantity of hydrogen leaving the stack is not feasible, as it would be expensive and would violate the economic constraints of the project. However, since the air fed to the burner is measured, and so is the entering natural gas flow and the stack temperature, it is possible to estimate the anodic λ with an enthalpy balance.

Assuming for now that natural gas is pure methane, with molar flow \dot{n}_{NG} and complete conversion to hydrogen and CO₂ according to reforming reaction 1 the flows entering the stack are:

$$\dot{n}_{\rm H_o}^{\rm in} = 4\,\dot{n}_{NG} \tag{3}$$

$$\dot{n}_{\rm CO_2}^{\rm in} = \dot{n}_{NG} \tag{4}$$

All the CO_2 will exit the stack unchanged, whereas hydrogen will be spent. Water vapour will also follow, and the flow is assumed to be saturated with it at the stack temperature:

$$\dot{n}_{\rm H_2}^{\rm out} = \dot{n}_{\rm H_2}^{\rm in} \, \frac{\lambda - 1}{\lambda} \tag{5}$$

$$\dot{n}_{\rm CO_2}^{\rm out} = \dot{n}_{\rm CO_2}^{\rm in} \tag{6}$$

$$\dot{n}_{\rm H_2O}^{\rm out} = \frac{y_{\rm H_2O}^{\rm sat}(T^{\rm stack})}{1 - y_{\rm H_2O}^{\rm sat}(T^{\rm stack})} \left(\dot{n}_{\rm H_2}^{\rm out} + \dot{n}_{\rm CO_2}^{\rm out}\right)$$
(7)

where y is the molar fraction in gas phase. The flow exiting the stack depends therefore on λ , stack temperature, and natural gas feed flow.

The enthalpy flow of the fuel mixture entering the burner can then be calculated by:

$$\dot{H}^{\text{fuel}} = \sum_{i} \dot{n}_{i}^{\text{out}} h_{i}(T^{\text{stack}})$$
(8)

where for convenience molar enthalpy h is calculated setting the reference to elements in their native state at standard conditions.

The enthalpy of feed air is similarly calculated, assuming a constant composition of $19\,\%$ oxygen and $81\,\%$ nitrogen:

$$\dot{H}^{\rm air} = 0.19 \, \dot{n}_{\rm air} \, h_{\rm O_2}(T^{\rm env}) + 0.81 \, \dot{n}_{\rm air} \, h_{\rm N_2}(T^{\rm env}) \qquad (9)$$

In the burner, the only reaction assumed to take place is the complete oxidation of hydrogen:

$$H_2 + \frac{1}{2}O_2 \longrightarrow H_2O \tag{10}$$

The exhaust flows are therefore:

$$\dot{n}_{\rm H_2}^{\rm ex} = 0 \tag{11}$$

$$\dot{n}_{O_2}^{ex} = 0.19 \, \dot{n}_{air} - 0.5 \, \dot{n}_{H2}^{out}$$
 (12)

$$\dot{n}_{\rm N_a}^{\rm ex} = 0.81 \, \dot{n}_{\rm air} \tag{13}$$

$$\dot{n}_{\mathrm{H}_{-}\mathrm{O}}^{\mathrm{ex}} = \dot{n}_{\mathrm{H}_{-}\mathrm{O}}^{\mathrm{out}} + \dot{n}_{\mathrm{H}_{-}}^{\mathrm{out}} \tag{14}$$

$$\dot{n}_{\rm CO_2}^{\rm ex} = \dot{n}_{\rm CO_2}^{\rm out} \tag{15}$$

And the exhaust enthalpy flow is given by:

$$\dot{H}^{\text{flame}} = \sum_{i} \dot{n}_{i}^{\text{ex}} h_{i}(T^{\text{flame}}) \tag{16}$$

Performing an enthalpy balance:

F

$$\dot{H}^{\text{fuel}}(T^{\text{stack}},\lambda) + \dot{H}^{\text{air}} = \dot{H}^{\text{flame}}(T^{\text{flame}},\lambda)$$
 (17)

where it is highlighted that both stack outlet and exhaust enthalpy flows depend on λ . Since all other variables required to calculate the enthalpy flows are measured, it is possible to solve the equation for λ with an iterative solver.

In this series of calculations, the weakest assumption is that natural gas is composed of pure methane; indeed, natural gas in the distribution grid can change its composition over the year depending on where the gas is imported from. The hypothesis of fully saturated flow from the stack is accurate during normal operation, but may not be correct during start-up. Molar flow measurements can be assumed to be reasonably accurate, since all gases are at low pressure and can be approximated as ideal gases. Temperature measurements are reliable, and because of the large difference between exhaust and stack temperatures the effect of noise is minimal.

The proposed state estimator can be used to monitor the current anodic λ without any additional sensors; if the estimated λ were to decrease below a minimum level, the current drawn from the stack can be immediately be reduced to avoid the risk of hydrogen starvation.

4. TESTS OF CONTROLLERS ON EMULATOR

To test the controllers proposed in the previous section, a system emulator has been programmed in LabVIEW. The emulator is not a full-fledged process simulator, but only a testbed to verify that the behaviour of the controllers corresponds to specifications. In particular, detailed simulations of some important aspects of the system's dynamics have been neglected, since the objective is to only verify the controller's operation, not to simulate the entire system. For example, CO adsorption transients were not modelled, since they are either too fast to be relevant (when increasing air bleed) or too slow to be detected (when decreasing).

The operation of the drying controller is exemplified in figure 2: when drying condition occurs, the pressure drop across the cathode loses its characteristic background noise. After an induction period of about 30 seconds, during which the estimator gradually changes its measurement of the noise level, the estimator eventually determines that a drying condition is in effect, and the controller reduces stack temperature to re-establish correct humidification.

In figure 3, a flooding event is detected when the noise in the voltage measurement exceeds the normal noise level

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Fig. 2. Emulated drying event and controller reaction.



Fig. 3. Emulated flooding event and controller reaction.



Fig. 4. Typical air bleed modification pattern.

expected in normal operation. Again, there is an induction period in which the estimator evaluates the condition before the controller increases temperature to increase evaporation.

With an emulated CO concentration of 10 ppm, the air bleed controller operates as shown in figure 4. After increasing from 0.8% (a minimum value for the controller) to 1%, the controller notices an improvement in voltage; it then proceeds to another increase to 1.2%, but this does not give any further improvement in stack performance, and it therefore decides to settle back to 0.9%, where it will remain until the next air-bleed test cycle.

The effect of a reduction in anodic λ from 1.4 to 1.2 are shown in figure 5. The reduction in λ is accompanied by a very strong reduction in flame temperature in the burner, which is therefore confirmed as a good indicator for the risk of hydrogen starvation. Note that burner temperature will likely be controlled in a real system in



Fig. 5. Effect of reduction of anodic λ from 1.4 to 1.2 on burner flame temperature (no change in air flow).

order to provide the correct temperature for the reformer, likely by manipulation of the burner air. In that case, the estimator will work just as fine, since the burner air inflow is also one of its inputs.

The various tests indicate that the controllers are able to detect and compensate the emulated degradation events, and whereas they are all operating at the same time, no interference appears to occur between them.

5. FUTURE WORK

The controllers have been synthesised and tested in an emulator, but will need to be tested on real systems before they are completely verified. In particular, it will be necessary to calibrate several control parameters that depend on the particular type of fuel cells and systems:

- Time horizon for pressure sampling for drying detection;
- Time horizon for voltage sampling for flooding detection;
- Amplitude of increase-decrease steps for air bleed and voltage sampling time;
- Nonidealities in the estimation of anodic λ , such as changes in the composition of natural gas, position of flame temperature sensor, etc.

Several of these tests are scheduled to be executed at the Centre for Solar and Hydrogen Energy Research of Ulm, Germany, during the summer of 2015 within the SAPPHIRE project. A final controller is scheduled to be implemented by the end of July 2015 and tested by early 2016.

5.1 Prognostics

In addition to the presented controllers, the SAPPHIRE project aims to counteract irreversible, long-term degradation of LT-PEMFCs by diagnosing their current state and using this information to calculate their Remaining Useful Life (RUL), which can then be optimised by the controller to find the best operating conditions (e.g. temperature, humidity) at which the stack should operate.

This work is currently ongoing at FCLAB, France, and is scheduled to be complete in the first half of 2015, after which the results will be integrated with the controllers presented here.

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6. CONCLUSIONS

Many phenomena can reduce the performance of LT-PEMFC stacks, but some can be counteracted by means of appropriate control measures.

An important factor in the marketability of μ CHP systems is their cost, and it is therefore not possible to include advanced sensors to obtain the most accurate and detailed picture of the system's internal state.

The presented approaches made use of measurements already present in Dantherm Power's μ CHP system (such as flame temperature, stack voltage) or new, but cheap and reliable sensors (stack temperature, pressure drop over cathode).

Tests with a LabVIEW emulator of the μ CHP system indicate that the approach is able to counteract reversible degradation phenomena and to estimate the anodic λ .

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