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SOFC 5 kW_e CHP field unit: effect of the methane dilution

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Abstract

TurboCare and Politecnico di Torino (Italy), have installed a SOFC laboratory in order to analyze the operation of two SOFC generators (Project EOS-100 kW and EBE-5 kW) built by Siemens Power Corporation (SPC). In the EBE project the installation of the SFC5 SOFC generator (3.5 kW_e and 3 kW_{th}) was carried out. To date, it has operated in the workshop canteen for more than 15,984 hours with very high reliability.

The real stack is a complex system not installed in a Lab environment, and has several effects of not-homogeneity in terms of electrochemical response to fuel or air management modifications. Moreover, many of the parameters of the stack are not directly measurable, and have to be inferred by indirect measurements.

In this paper, the analysis of the not homogeneous behaviour of the different segments of the complete stack is performed, through an experimental session using a not conventional fuel.

The obtained data have been analyzed using the ANOVA for every dependent variable and a non-linear regression model for the voltage. Those models were used to evaluate the effect of the fuel modification on the local fuel utilization in different sectors of the stack. Keywords: SOFC, real stack, diluted fuel, factorial design.

1. Introduction

TurboCare S.p.A. (TC) and Politecnico di Torino, both located in Torino (Italy), have installed a SOFC laboratory in order to analyze the operation, in a cogenerative configuration, of two generators built by Siemens Power Corporation (SPC): the SOFC CHP100 and the 5 kW Alpha6 Generator.

Some experimental and modeling activities were performed during the four years of operations, involving some aspect of installation and operation in a real industrial environment.

The EBE project (Energia a Basse Emissioni) concern the Alpha6 Generator (SFC5 alpha6), providing 3.5 kW_e of electric energy delivered to the grid and about 3 kW_{th} of thermal energy supplied to the TC canteen. The generator was installed in April 2006, and at present it runs from more than 15,900 hours and has delivered to the grid about 47.7 MWh. The initiative is highly innovative because in Italy there are not plants for micro-cogeneration on 1-5 kW scale based on solid oxide fuel cell. This technology is devoted to small electrical generation for civil use (big flat, small office, house).

The Politecnico di Torino developed several activities in the laboratory [1-6]. In [7] a sensitivity analysis of voltages against fuel utilization and generator temperature, and an optimization of the system by regression model, has been proposed, while another experimental campaign was devoted to study the effect of air distribution inside the stack. Then an experimental campaign has been carried out in order to analyze the effect of

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changing the most important operating parameters (generator temperature, fuel utilization and current), in order to obtain the generator polarization model [8].

The real stack is a complex system not installed in a Lab environment, and has several effects of not-homogeneity in terms of electrochemical response to fuel or air management modifications. Moreover, many of the parameters of the stack are not directly measurable, and have to be inferred by indirect measurements.

Compared to studies on a single cell, where the temperature can be easily controlled to impose homogeneity on the cell surface, on a large SOFC generator we have different temperature profiles on cells placed in different segments of the stack. In particular, it is possible to observe that for the segments close to the in-stack reformer the temperature is lower than the other part of the generator (due to the cooling effect of the steam reforming reaction occurring into the reformer section). Another cause of this not homogeneity of temperature distribution is due to the fluid-dynamic of the reactant flows inside the stack (air and fuel) [7].

Moreover, a further problem on the characterization of large systems is the difference of pedigree and degradation level on the single cells that compose the stack [9].

Biogas fuel feeding present an attractive option among emerging applications for fuel cells, especially for solid oxide fuel cell. Usually, biogas is a local resource and residues from farms and municipalities typically represent small fuel sources (in the range of 5-100 kW) suitable for fuel cell system.

Several groups have studied SOFC fuel flexibility in recent years using theoretical analysis or simulation work [10-14]. A 1 kW_{el} SOFC unit from Sulzer HEXIS, Switzerland, was successfully operated on farm biogas in Switzerland for one year [15,16]. Fuel Cell

Technologies (FCT) from Canada demonstrate 5 kW_{el} SOFC units also for biogas application [17].

In this paper, the analysis of the not homogeneous behaviour of the different segments of the complete Siemens 5 kW alpha6 stack is performed, through an experimental session using a not conventional fuel. The scheduled tests have been carried out in April 2008. This is the first experimental campaign using a non conventional fuel and it yelded interesting results about fuel distribution inside the stack, which are presented in section 8.

2. System description

The Alpha6 Generator utilizes the commercial prototype air electrode supported cells (22 mm diameter, 75 cm active length, 384 cm² active area). The generator is fed with natural gas from the grid distribution. In Figure 1 the picture representing the Alpha6 Generator test site in TurboCare is shown.

Figure 1. Picture of the Alpha6 Generator test site in TurboCare (Torino, Italy)

The upper level of system hierarchy after the single cell is the sector, which consists of a 22-cell array arranged as 11 cells in electrical series by 2 cells in electrical parallel; 4 sectors are aligned side by side, interconnected in serpentine (for a total of 88 single tubes). In Figure 2 a simplified scheme of the Balance of Plant of the 5 kW_e Alpha6 Generator is shown.

Figure 2. Simplified scheme of the Balance of Plant of the 5 kWe Alpha6 Generator

The SOFC module includes an in-stack fuel reformer which is placed in the middle of the stack. This reformer uses heat from the SOFC operation to provide the energy needed to convert the hydrocarbon fuel into hydrogen and carbon monoxide. The SOFC module also includes an internal air recuperator to recover heat allowing higher operational efficiency. The function of the ECS (Electronic Control System) is to monitor all the components and sensors (current, voltage, temperature, etc.) in addition to load demands from the inverter. In the following, some results obtained in different previous experimental campaigns for sector voltages, system AC power and efficiency are presented [8]. Experimental data have been analyzed using the design of experiment approach, which allows to apply analysis of variance (ANOVA) [18] and to obtain a regression model for the dependent variables. All the results presented in this section derive from the analysis of the regression models. In Figure 3 the cell polarization for sector A and sector B, at fixed average stack temperature (T_{gen} =960°C), and as a function of fuel utilization (FU), are shown.

Figure 3. Polarization model for sector A and B, function of FU

Voltages have an expected behavior when varying fuel utilization parameter. In both sectors it is evident that voltage sensitivity to FU is higher at high FU values: at high FU, a variation of FU corresponds to a larger variation in the cell voltage, with respect to the low FU case. In sector B it is also possible to note larger fuel sensitivity at high current density. In Figure 4, AC power output of the system for different T_{gen} and FU conditions is shown.

Figure 4. AC power model, function of current density, T_{gen} and FU

AC electric power has an expected behavior: an increase in generator average temperature causes an increase in AC power, and this fact is mainly due to a reduction of ohmic losses. The higher temperature causes also a reduction of the activation overvoltages. Besides, an increase in fuel utilization causes a reduction of the AC power. This behaviour is mainly due to Nernst voltage dependence on anodic partial pressures.

In Figure 5 the behavior of the global efficiency, with variations of T_{gen} and FU, is shown.

Figure 5. Global efficiency model, function of current density, T_{gen} and FU

With lower generator average temperature, it is observed a slightly higher global efficiency. This fact can be interpreted considering that the global efficiency is the sum of the AC electrical efficiency and of the thermal efficiency. In this case, the decrease in AC efficiency, due to larger ohmic losses, is balanced by the increase of thermal efficiency. The latter is linked to a higher air flow (needed for the stack cooling because of the larger ohmic losses), which makes available a higher heat recovery.

It is interesting to analyse the variations of global efficiency with fuel utilization. It can be seen from Figure 5 that exists a value of fuel utilization, within the experimental domain, that maximizes the global efficiency. It follows directly from the observation that AC efficiency is monotonic w.r.t. FU, while thermal efficiency presents a maximum at a certain FU value. The latter is due to a balance between the effects of FU on two parameters: 1) exhaust mass flow; 2) exhaust temperature.

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Rising FU results in an enhancement in heat production into the stack, due to an increase in irreversibilities and a reduction in the cooling effect of the fuel flow, and this causes an increase of the air flow (and consequently of the exhaust flow) in order to control the average stack temperature; but the prevailing effect is a reduction of the exhaust temperature (due to the reduction of the heat from the post-combustion), causing a reduction of the heat recovered. Reducing FU the increase of the stack translates in a reduction of the heat from irreversibility, and a decrease of the air (and exhaust) mass flow. Therefore, there is a value of the FU where a balance of the two effects occurs, causing a maximization of the thermal recovery and as consequence a maximization of the global efficiency.

An important feature of SFC 5 alpha6 is the possibility to control the air flowing to every single sector of the stack. The air management system consists in a series of manual valves that allow to control the air flow through the single sectors of the stack. An experimental campaign has been carried out in order to evaluate the influence of the air management system on the system performance and to find the optimized configuration [7]. After this experimental campaign, every other experimental test has been performed with the optimized configuration of air management system.

3. Description of the experimental session

The scheduled tests have been carried out in April 2008, with the aim to characterize the system with different fuel compositions, different loads (current) and generator setup (average) temperature.

Besides, we wanted to demonstrate the possibility to operate the system when fed with a fuel with low LHV. This fuel is obtained introducing in the main natural gas stream a flow of NHmix (5% of Hydrogen in Nitrogen). In this way a methane dilution is achieved. In the following we will refer to this diluted mixture as "diluted fuel" (see Table 1). The choice to use NHmix was linked to very practical reasons: this mixture is used during the start up and shut down of the stack, and all the connections with the stack are already placed in the BoP.

Moreover, the variation of the Fuel Utilization factor allowed to perform a sensitivity analysis of voltages against fuel utilization. In fact, as already discussed and performed in [22], the fuel sensitivity tests can be used as a diagnostic tool to investigate the fuel distribution on a large SOFC stack. As discussed below, an interesting result has been obtained: when the system is fed with this diluted fuel, a better fuel distribution inside the stack has been obtained, if compared to simple natural gas feeding.

The test session has been planned following a factorial experimental design, where some controllable variables (factors) are varied among discrete possible values or "levels" (usually two: low and high value); the complete experiment takes on all the possible combinations of these levels across all those factors [18], allowing the analysis of the main and interaction effects of all the factors on the dependent variables of the system. Regression models of different orders, expressing the functional relation between factors and dependent variables, can be obtained. In our session, the factorial design has been used

to evaluate the main and interaction effect of the three factors (generator average temperature (T_{gen}), stack current (I) and fuel composition – see Table 1) on several dependent variables expressing the performance of the generator. The design of experiment adopted is a simple 2³ factorial design (Figure 6), which allows to obtain first order regression model of the dependent variables as functions of the independent factors. The range of variation of each factor has been imposed in agreement with TurboCare, in order to avoid malfunction of the generator.

Figure 6. 2³ factorial design

Table 1. Values of the factors ion the factorial design

In Table 2 the fuels composition in the two considered fuel options (natural gas and diluted fuel) are shown.

Table 2. Fuels composition of the experimental session

It has to be specified that the thermal balance of the complete stack can be influenced by the chemical reactions occurring in the "in-stack" reformer, where different reactions (exothermic and endothermic) take place whether fuels of different composition are sent to the reformer unit. The different enthalpies of reaction could determine a modification of the thermal balance, and thus of the average stack temperature: as an example, since the steam-reforming of a ethanol stream is somewhat less endothermic than the methane one, a higher stack temperature could be observed. Anyway, in these SOFC systems the stack temperature can be considered

as a independent variable (thus not depending on the reactions occurring in the reformer), since the air stoichs is the parameter managing the stack temperature: we could impose a air stoichs in such a way to keep the stack temperature constant. When a less endothermic reforming reaction is taking place, an increase of the the stack temperature is avoided augmenting the air flow into the stack in order to cool down the Generator.

4. ASR data analysis

A preliminary analysis on the sector voltage data consisted in the evaluation of the Area Specific Resistance (ASR). ASR is an index of the slope of the polarization curve that can be used as an indicator of the performance of the cells. It is defined by dV/di at 0.7 V and is determined from the slope of the best fitting line over the measured data within the interval 0.65 - 0.75 V. In our case it is not possible to evaluate the ASR with the given definition because the tests have been designed with only two current levels, thus it is evaluated as:

$$ASR = \left| \frac{V1 - V2}{i1 - i2} \right|$$
(1)

In Figure 7 the influence of generator setup temperature on sectors ASR with different fuel is shown.

Figure 7. ASR distribution function of fuel composition and generator average temperature

In both cases an increase of the generator average temperature causes a reduction of the ASR. All the overvoltages of the polarization curve are influenced by the generator average temperature but the ohmic overvoltage is the most influenced by this parameter, and it decreases with an increase of the generator average temperature. The variations of ASR with temperature is more evident on sectors B.

From the figure it is evident a different slope for the sectors. This is due to higher ohmic losses in sector B, caused probably by the partial interconnection detachment and a consequent increase in contact resistance.

In Figure 8 the average ASR comparison for different feeding fuel is shown.

Figure 8. ASR function of the fuel composition

When the system is fed with diluted gas, the ASR of each sector decreases. This behaviour cannot be explained with a temperature effect (on the contrary: the diluted fuel causes even a decrease of the local temperature); therefore, this positive effect can be linked probably to a fluid dynamic effect, which is an apparent better fuel distribution inside the stack. This effect will be analysed and explained later in section 8.

5. Regression model

This experimental campaign has been carried out following the factorial design in Figure 2, in order to apply the ANOVA model and to study the effect of each parameter independently. Nevertheless, the system has not been able to keep sufficiently stable operation conditions in terms of fuel utilization, a parameter with an important influence on the voltage and which has not been considered as a factor in the design of experiment. This

inconvenience is caused by the control system. In fact, SFC5 is a pre-commercial prototype and the actual control system is designed for equipment safety purposes and is not suitable for thorough experimentation. To overcome this problem and to allow more accurate testing sessions, it would be necessary to develop a dedicated control system. Because of the difficulties explained, we couldn't rely on the ANOVA analysis for our data, and decided to perform a non-linear fit on the voltages, as functions of FU, generator average temperature and current density. We treated separately the data for different fuel conditions: so different regressions model of the voltage for natural gas and diluted gas, namely V_{NG} and V_{SG} , were obtained.

Due to the complexity of the system, we decided to not apply a typical polarization model. In fact in a large stack, like the Alpha6 Generator, there are some factors that cannot be perfectly under control, i.e. local fuel distribution or local temperature. For this reason we decided to adopt a more empirical model, although inspired by the standard analytical polarization model for single cells. In particular, we considered Nernst, ohmic, activation and concentration (i.e. depletion of reactants) overvoltages. Only the effect of diffusive transport in the porous anode and cathode has not been considered in the model because, in the operating conditions (low-medium current densities), it resulted negligible compared to the others.

The classical expression for reversible voltage is [19]:

$$V_{rev} = -\frac{\Delta G^R(T)}{2F} + \frac{RT}{2F} \log\left(\frac{P_{H_2} P_{O_2}^{0.5}}{P_{H_2O}}\right)$$
(2)

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The partial pressure dependence of this expression can be rewritten in terms of Fuel Utilization factor and air stoichs, both measured by the control system [22]:

$$\dot{\mathbf{E}} = \dot{\mathbf{E}}_{0} - \frac{\mathbf{R} \cdot \mathbf{T}}{2 \cdot \mathbf{F}} \cdot \ln \left[\frac{\mathbf{FU} \cdot \left(\frac{\lambda}{0.2 \pi} - \mathbf{FU} \right)^{0.5}}{(1 - \mathbf{FU}) \cdot (\lambda - \mathbf{FU})^{0.5}} \right]$$
(3)

As a further simplification, temperature dependence is neglected due to the not significant effect linked to the low temperature modification in the tests, and a simple log(1-FU) term is used to take into account the dependence on the partial pressures of gases.

Concerning Ohmic overvoltage, it is linear with respect to the current, and a simplified empirical dependence on temperature has been assumed. Activation overvoltage was modeled by means of the standard Butler-Volmer expression, again neglecting temperature dependence. The following non-linear expression has been chosen for the voltages:

$$V(FU, T, i) = \underbrace{\beta_1}_{\text{Nernst}} + \underbrace{\beta_2 \log(1 - FU)}_{\text{Concentration}} + \underbrace{\beta_3 \frac{i}{\log \frac{1}{T}}}_{\text{Ohmic}} + \underbrace{\beta_4 \sinh^{-1} \left[\frac{i}{\beta_5 FU \exp \frac{-10^5}{FU}}\right]}_{\text{Activation}}$$
(4)

A remark should be added about β_5 : due to the great discrepancy of literature data regarding exchange current density for the cathode, we chose to add this fitting parameter. The best fitting values of the parameters β_i have been found with the Gauss-Newton method [20]. As seen, we have very simple temperature dependence, involving only one parameter. On the other hand the FU appears in two terms and three parameters can be tuned in order to fit accurately the model to the data variations.

Since the expression chosen is suggested by physical considerations but represents a simplified empirical model, the β_i values don't have a strong physical meaning. For the other dependent variables a linear model building procedure is adopted

6. Voltage analysis

In Figure 9 the effects of current, fuel utilization and fuel composition on the sector voltages obtained with the regression model are shown, and compared with the experimental data.

The difference between regression model and experimental data are due to the fact that the model can fix the values of independent factors (essentially, setup temperature and FU), while in the real experimental session the control system is not able to maintain a constant value at the desired set point but there is a tolerance in the values of setup temperature and FU.

Figure 9. Polarization regression model function of current density, FU and fuel composition

Sector A shows an expected behavior. In fact, when the system is fed with diluted fuel, a voltage decrease is observed, due to the dependence of the exchange current density on reactant partial pressures. Also an increase in fuel utilization causes a voltage decrease. On the other hand, sector B shows an unexpected behavior with respect to variations in the feeding fuel. Here, when the system is fed with diluted fuel, the voltage is higher than with

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natural gas feeding. We will discuss this topic in the following section, with the aid of sensitivity analysis.

The developed regression model operates quite well and also confirms the same observation made experimentally: a different slope for sector B due to higher ohmic losses, caused probably partial interconnection detachment and a consequent increase in contact resistance.

8. Fuel sensitivity and local fuel utilization estimation

In this paragraph the voltage sensitivity to fuel utilization and its relation with local fuel utilization are presented. Before focusing on the topic, it must be observed that the FU parameter is a global variable, i.e. we don't have different measurements for each sector but only one for the whole stack.

Sensitivity of voltage to fuel utilization (dV/dFU) is an important variable when studying solid oxide fuel cell systems. In a previous work [21-22] it has been demonstrated that the sensitivity of cell voltage to fuel utilization depends on several contributions, which concern the Nernstian term, the diffusion term and eventually the effect of leakages of air at the anode side. At low current density the fuel utilization sensitivity is governed essentially by the variation of the average Nernst voltage with FU whereas at high current density the effect of diffusion and leakages become more effective. Moreover, it has been shown that there is a direct relationship between the voltage sensitivity to fuel utilization and the local value of the fuel utilization in operation, and therefore to the fuel distribution inside the stack.

Considering the obtained regression model of the sector voltage, it is possible to calculate analytical expressions for the derivative (dV/dFU), as a function of Fuel Utilization. The local fuel utilization of a sector can be estimated by comparing the measured fuel sensitivity value (measured value F^0) with the expression calculated from the voltage regression model. In other words, we look for a solution of:

$$\frac{\partial V(FU)}{\partial FU} = F^0 \tag{5}$$

The solution is the estimation of local FU. Note that here the only variable is FU, since temperature and current are measured and inserted into the regression model.

This procedure can be performed both with natural gas and diluted fuel, therefore allowing an analysis of the fuel distribution in the stack sectors with different fuel conditions.

The distribution of fuel utilization sensitivity, according to the proposed model, depends on the local temperature and on the local fuel utilization; a limit of the estimation is that the effect of the temperature is neglected in the following analysis, due to the fact that we cannot know the local temperature.

In Figure 10 it is shown how local FU values are obtained from voltage sensitivity curves for sectors A and B.

It is well known that differences between local fuel utilization values are caused by a non uniform fuel distribution among the sectors: a higher local FU means a lower fuel flow to the sector.

Figure 10. Estimation of local values of fuel utilization through analysis of voltage sensitivity to fuel utilization

With diluted fuel, the sensitivities are much higher than with natural gas. The evaluation of local fuel utilization in Table 3 compares two sectors in two different feeding conditions.

Table 3. Estimation of local fuel utilization

With natural gas, sector B shows a higher value of local FU, so it seems that sector B is penalized by the fuel distribution. On the other hand, when the system is fed with diluted fuel a higher mass flow reaches the anode side of the sector B. In these conditions sector A shows a local FU increase and a slight voltage decrease (Figure 4), while sector B shows an opposite behavior. It seems likely to say that with diluted fuel feeding the anodic molar flow is nearly double, and for this reason the fuel is better distributed inside the stack. Sector B, which is penalized in normal conditions because of a bad fuel distribution, can take advantage of the increased anodic flow. It is important to take into account also the role of the leakages of air to the anode side. In fact, if air leakage is large we have an important quantity of hydrogen that burns in the stack, with the consequence that it does not take part in electrochemical reactions generating current. If this occur, the effective operational local fuel utilization is higher than the expected one and sensitivity to the fuel is supposed to be very high. When feeding the system with a diluted gas, the anodic pressure increases and the leakages are reduced.

These important results are confirmed by the study of electrical behavior of the stack. In particular, it is possible to observe that when the system is fed with diluted fuel the

polarization curve of the sector B is better than with natural gas, as noticed in the ASR results (Figure 8).

9. Conclusions

In the paper, the experimental campaign had the aim to demonstrate the capability of the system to run with low LHV fuel, and especially to analyse the complex effects of feeding a diluted fuel on a large stack composed of several sectors. The results can be summarized as follow:

- Was demonstrated the capability of SOFC stack to operate with low energy content in the fuel.
- The ASR of the different sectors decreases when the system is fed with diluted fuel: this behavior is caused by the better fuel distribution inside the stack (at the same condition activation overpotential decreases).
- The ASR value of Sector B is higher than other sectors: this behavior is probably due to partial interconnection detachment that causes an increase of ohmic overvoltage.
- A simplified not linear regression model starting from the physical expression of cell voltage was developed in order to investigate the experimental domain.
- Sector A and sector B voltages show opposite behaviors when the system is fed with diluted fuel: sector A shows a voltage decrease while sector B shows a voltage increase.

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- The analysis of the voltage sensitivity to fuel utilization can be used as a diagnostic tool to estimate the local distribution of the fuel utilization in the different sectors of the stack
- Using the diluted fuel the voltage sensitivities to fuel utilization are much higher than using natural gas.
- With natural gas feeding sector B shows a higher local FU than sector A. With diluted fuel sector A shows a local FU increase and a slight voltage decrease, while sector B shows an opposite behavior.
- With diluted fuel feeding, the fuel is better distributed inside the stack: therefore, the dilution causes an increase of the performances of the whole stack, because it determines a better internal fluid dynamic

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5	FIGURE CAPTIONS
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Table	1. V	alues	of the	factors	ion	the	factorial	design
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Factor	Min	Max
Diluted fuel	Natural Gas	Diluted Fuel
T _{gen} (°C)	949.6	967.8
I (A/cm ²)	0.109	0.137

 Table 2. Fuels	s compositi	on of the	experimenta	al session

Gas composition	C _x H _y (%) (hydrocarbons, mainly methane)	H ₂ (%)	CO ₂ (%)	N ₂ (%)
Natural gas	97.162	0.000	1.106	1.732
Diluted fuel	48.770	2.490	0.555	48.185

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Table 3. Estimation of local fuel utilization

	Voltage sensitivity to FU	Local FU
	[mVcell/%FU]	
Sector A – Natural gas	-1.09	0.807
Sector A – Diluted fuel	-3.54	0.812
Sector B – Natural gas	-2.73	0.861
Sector B – Diluted fuel	-4.09	0.746



Figure 1. Picture of the Alpha6 Generator test site in TurboCare (Torino, Italy)





Figure 2. Simplified scheme of the Balance of Plant of the 5 kWe Alpha6 Generator



Figure 3. Polarization model for sector A and B, function of FU





Figure 4. AC power model, function of current density, T_{gen} and FU



















Figure 8. ASR function of the fuel composition



Figure 9. Polarization regression model function of current density, FU and fuel composition





Figure 10. Estimation of local values of fuel utilization through analysis of voltage sensitivity to fuel utilization