Electric Field Modifier Design and Implementation for Transient PEM Fuel Cell Control

MARC SCHUMANN, FLORIAN GRUMM, JAN FRIEDRICH, DETLEF SCHULZ
Electrical Power Systems
Helmut Schmidt University / University of the Bundeswehr Hamburg
Holstenhofweg 85, 22043 Hamburg
GERMANY
marc.schumann@hsu-hh.de https://www.hsu-hh.de/ees

Abstract: - Common controlling strategies of fuel cells regulate parameters like the flow rates, pressures, temperatures, and relative humidities of the supplied gases. These strategies have a slow control effect on the fuel cell output voltage, especially at high dynamic loads, which is why fuel cell voltage and power drops for several seconds to minutes after a load step. Today, an oversizing of fuel cell systems is necessary to meet the requirements of dynamic load profiles. This paper deals with the design and implementation of an electric field modifier (EFM) control unit into fuel cells to enable the regulation of an additional control parameter, which is considerably faster than the common parameters. The EFM control unit consists of EFM electrodes that are placed directly on or in the membrane of polymer exchange membrane fuel cells and are connected to an external controllable voltage source. Possible electrical connections and actuating signals are presented. Deduced advantages include a better dynamic fuel cell system voltage behavior, a cost- and weight-optimized on-board grid integration, and a prolonged membrane durability.

Key-Words: - electric field modifier design, electric field modifier implementation, electrically controllable membrane electrode assembly, dynamic behavior, electrical connection, actuating signals

1 Introduction

For a successful change of the energy supply towards a supply with a high share of renewable energies, a coupling of the energy sectors is required. Proton exchange membrane (PEM) fuel cells are a promising technology for this, because they have a quick start and shutdown performance due to low operating temperatures of 60 – 80 °C. Moreover, they are able to use nearly the complete partial load range and are characterized by relative high current densities.

In Fig. 1 a), a typical characteristic *I*-*V*-curve and *I*-*P*-curve of a fuel cell is depicted. Voltage is decreasing with higher currents due to different voltage losses, which are explained in Section 2.1.

Actual challenges of fuel cells for usage in applications with frequent load changes include dynamic behavior of voltage and power. Slow fuel cell system dynamics lead to variations in voltage after a current step [1] – [4]. Hence, fuel cell voltage fluctuates for quite a few seconds before reaching a new steady-state operation point on the curve depicted in Fig. 1 a). Fig. 1 b) shows this dynamic voltage behavior of fuel cells for a voltage drop. Curve b.1 presents an ideal voltage response, where voltage follows directly the current step. Due to transient and dynamic phenomena described in Section 2.1, real fuel cells display a voltage characteristic as shown in curve b.2. Many research groups have developed different controlling strategies up to now [5], but none of these is fast enough to compensate fully the slow dynamics of fuel cells. As a result, fast responding components, like batteries or super capacitors, are used to compensate this voltage drop in on-board power supplies [6] – [8], which leads to high integration costs and system weights.

In addition, membrane durability is still a technical challenge in actual PEM fuel cells. Low durability leads to frequent maintenance repairs and increases costs. There are several reasons for current durability problems. One of these is an inhomogeneous distribution of reactants, which leads to locally higher currents, and therefore to local temperature hot spots damaging the membrane [9].

This contribution presents the design and implementation of a new controlling method of PEM fuel cells using an additional electric field modifier (EFM) control unit. The paper is structured as follows: Section 2 presents a mathematical description of transient fuel cell output voltage. In Section 3, the concept, design and implementation of the EFM control unit as a new way of controlling

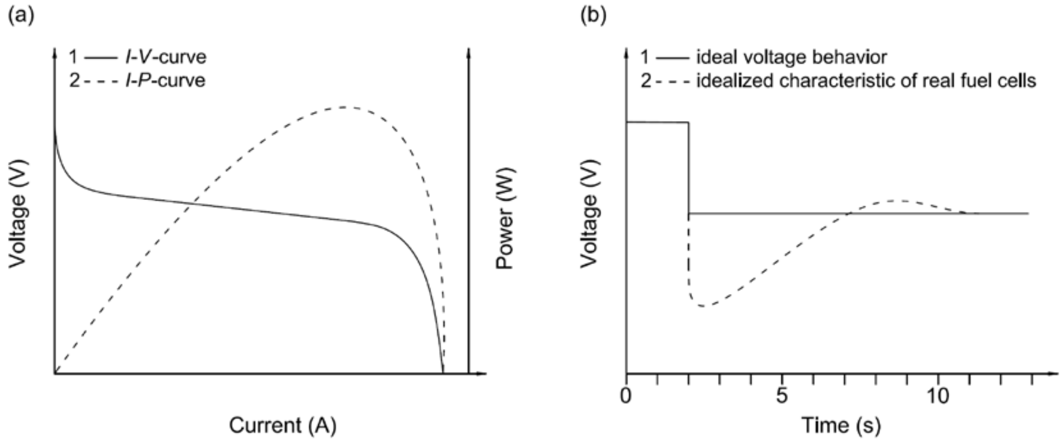

Fig. 1. a) Characteristic *I-V*-curve (a.1) and *I-P*-curve (a.2) of a fuel cell; b) Dynamic fuel cell voltage characteristics after a positive current step at $t = 2$ s: b.1) ideal voltage response; b.2) idealized dynamic voltage behaviour of actual fuel cells.

PEM fuel cells is presented. Subsequently, a discussion of expected advantages due to the usage of an EFM control unit is given in Section 4. Finally, Section 5 draws a conclusion on electrically controllable PEM fuel cells with an EFM control unit.

2 Mathematical Description of Fuel Cell Voltage

2.1 Computation of Fuel Cell Voltage

Reversible fuel cell voltage under non-standard-state conditions E_{Nernst} is given by (1)

$$E_{\mathrm{Nernst}} = E^0_{\mathrm{Nernst}} + \frac{\Delta s}{nF}(T - T_0) - \frac{RT}{nF} \cdot \ln \frac{\prod a^{v_i}_{\mathrm{products}}}{\prod a^{v_i}_{\mathrm{reactants}}} \tag{1}$$

with the standard-state Nernst voltage E^0_{Nernst}, molar entropy s, number of electrons transferred in the reaction n, Faraday constant F, operating temperature T and standard-state temperature T_0, ideal gas constant R, and chemical activity a of reactants and products with their respective stoichiometric coefficient v_i be taken into account.

In reality, steady-state voltage V_{fc} of fuel cells decreases with increasing current and can be expressed with the following equation:

$$V_{\mathrm{fc}} = E_{\mathrm{Nernst}} - V_{\mathrm{act}} - V_{\mathrm{ohm}} - V_{\mathrm{conc}} \tag{2}$$

where V_{act}, V_{ohm} and V_{conc} are the three different voltage losses in fuel cells. Activation losses V_{act} due to slow reaction kinetics dominate at low currents. Electrical and ionic conduction lead to ohmic losses V_{ohm} represented by a nearly linear voltage drop. At high currents, fuel cell voltage is limited because of reactant depletion at the reaction sites, expressed by concentration losses V_{conc}.

After a change of power demand, fuel cell voltage changes according to the characteristic *I-V*-curve in Fig. 1 a). It takes some time for the voltage to reach the next steady-state operation point, compare Fig. 1 b). The reason for this are different phenomena occurring in fuel cell systems. In Tab. 1, important transient and dynamic effects and their effective time range are presented. The fastest phenomenon with a time scale of less than a second is the electrochemical double layer, which exists between the electrodes and the membrane. This is followed by a delay of gas diffusion to the reaction sites in the range of one to two seconds. In a time range of ten to 100 seconds or more, removal of excess water at the cathode, and control of membrane humidification and operational temperature leads to voltage deviations, before voltage is stabilizing at the new operation point [1], [3].

Table 1 Classification according to the effective time range of transient and dynamic effects in fuel cells after a current step [1].

Transient / Dynamic Effect	**Time Range**
Electrochemical double layer	< 1 s
Gas diffusion	1 - 2 s
Membrane humidification and cathode flooding	> 10 s
Variation in temperature	> 10 s

Fig. 2 a) shows a simple equivalent circuit model based on [10] to describe transient fuel cell voltage. Herein, resistors are used to model the voltage losses. A capacity C_{dl} is connected in parallel to the resistors representing the activation and concentration losses to take the effects of the electrochemical double layer into account. Transient voltage can be calculated with (3)

$$\frac{\mathrm{d}v_{\mathrm{d}}}{\mathrm{d}t} = \frac{i}{C_{\mathrm{dl}}} - \frac{v_{\mathrm{d}}}{\tau} \tag{3}$$

where v_{d} is the voltage drop across $R_{\mathrm{act}} + R_{\mathrm{conc}}$ and τ is the time constant of the equivalent circuit. In the event of a negative current step, τ can be determined with (4) [10]:

$$\tau_{\mathrm{negStep}} = C_{\mathrm{dl}} \cdot (R_{\mathrm{act}} + R_{\mathrm{conc}}) = \frac{C_{\mathrm{dl}} \cdot (V_{\mathrm{act}} + V_{\mathrm{conc}})}{i_{\mathrm{fc}} - i_C} \tag{4}$$

where i_{fc} is the fuel cell current and i_C is the capacitor current, see Fig. 2 a).

In opposition to [10], τ has to be calculated with (5) in the event of a positive current step, which can be derived from the equivalent circuit diagram.

$$\tau_{\mathrm{posStep}} = C_{\mathrm{dl}} \cdot R_{\mathrm{ohm}} = \frac{C_{\mathrm{dl}} \cdot V_{\mathrm{ohm}}}{i_{\mathrm{fc}}} \tag{5}$$

This indicates a decrease of transient effects of the electrochemical double layer for a positive current step, when ohmic losses are minimized. Finally, transient fuel cell voltage $v_{\mathrm{fc,trans}}$ behavior can be computed with (6).

$$v_{\mathrm{fc,trans}} = E_{\mathrm{Nernst}} - V_{\mathrm{ohm}} - v_{\mathrm{d}} \tag{6}$$

2.2 Mathematical Extension for electrically controlled PEM Fuel Cells

In PEM fuel cells, electrical and chemical driving forces lead to proton conductivity through the membrane. Mainly responsible are electrical driving forces due to a voltage gradient across the membrane [11]. Charge flux because of a voltage gradient j_{voltage} can be determined with (7) [11]:

$$j_{\mathrm{voltage}} = \sigma \cdot \frac{\mathrm{d}V}{\mathrm{d}x} = \sigma \cdot \frac{V}{d_{\mathrm{membrane}}} \tag{7}$$

where σ is the ion conductivity, V is the applied voltage, and d_{membrane} is the thickness of the membrane. Standard electrode potential in H_2O_2 fuel cells is ca. 1.23 V. It can be assumed, that this electrode potential is equal to the applied voltage at the membrane if voltage losses are not considered.

To control directly the voltage behavior after a current step, the effect of voltage gradient on the proton transport will be used by controlling the voltage gradient at the membrane. For this reason, an electric field modifier (EFM) is integrated into the current fuel cell setup to generate and control the applied voltage at the membrane [12] – [15]. This leads to a temporary adjustment of ohmic losses and thus, to the possibility of directly controlling proton conductivity. With this, the general mathematical description of transient fuel cell voltage for electrical controllable fuel cells is

$$v_{\mathrm{fc,trans}} = E_{\mathrm{Nernst}} - V_{\mathrm{ohm}} - v_{\mathrm{d}} \pm v_{\mathrm{e-control}} \tag{8}$$

where $v_{\mathrm{e-control}}$ is assumed to equal the applied voltage at the membrane in a first approximation. $v_{\mathrm{e-control}}$ can be positive or negative, depending on whether voltage amplification or damping is needed. Fig. 2 b) expands Fig. 2 a) by the EFM control unit. Shortly conducted experimental investigations will help to analyze $v_{\mathrm{e-control}}$ in order to understand the

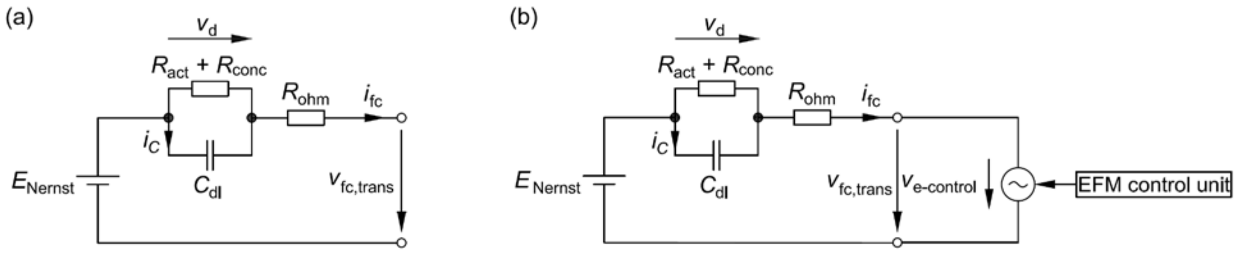

Fig. 2 a) Equivalent circuit model of fuel cells, based on [10]: Non-standard-state Nernst voltage E_{Nernst}, transient voltage drop v_{d} across activation and concentration losses represented by a resistor $R_{\mathrm{act}} + R_{\mathrm{conc}}$, ohmic losses represented by a resistor R_{ohm}, double layer capacitance C_{dl}, capacitance current i_C, fuel cell current i_{fc}, and transient fuel cell voltage $v_{\mathrm{fc,trans}}$; b) New equivalent circuit model of fuel cells with an additional electric field modifier (EFM) control unit: Internal controlling voltage $v_{\mathrm{e-control}}$.

effects of electrically controlled transient voltage.

3 Electric Field Modifier Control Unit

3.1 Design and Implementation of the Electric Field Modifier

Actual PEM fuel cell configurations need an adjustment to generate the additional electric field at the membrane. To achieve this, EFM electrodes are integrated into the setup to enable controllability. With this, the typical membrane electrode assembly (MEA) of a PEM fuel cell will be upgraded to an electrically controllable MEA (EC-MEA). EFM electrodes can be realized in various ways; some examples of possible grid structures are depicted in Fig. 3. Fig. 3 a) – c) show different types of single controlled EFM electrodes. Fig. 3 d) depicts an example of a single comb-shaped structure, whereas Fig. 3 e) illustrates a double controlled and intertwined comb-shaped structure to enable a current density homogenization. In Fig. 3 f) a circle pattern is shown. Generally, all imaginable structures to effectively generate and control an additional electric field and to homogenize current and temperature density are possible.

Furthermore, there are different possibilities to integrate EFM electrodes into the fuel cell setup. Potential configurations of EC-MEA include single and multi EFM electrode setups. In Fig. 4 and Fig. 5 four examples of how to integrate the EFM electrodes are shown, but more combinations are possible. Fig. 4 depicts two examples of a single EFM electrode in the middle of the membrane, but it is also possible to install the electrode between the membrane and the catalyst layer. In Fig. 4 a) and Fig. 5 a), the potential of the EFM electrodes is independent of the fuel cell electrode potentials. In the case of the single EFM electrode in Fig. 4 a), the counter potential is outside of our fuel cell system, for example the ground. Fig. 4 b) and Fig. 5 b) show examples of EFM electrodes, where the potential is dependent on one of the fuel cell electrode potentials. It could be the anode as well as the cathode potential. A variable DC or AC voltage can be used to actuate the EFM electrodes. The EFM electrodes could be printed on the outside of the membrane or be integrated in the membrane during the manufacturing process.

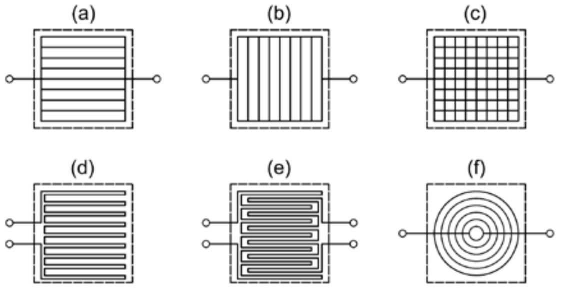

Fig. 3. Simplified drawings of possible single and double layer electric field modifier (EFM) electrodes: a) horizontal grid; b) vertical grid; c) meshed grid; d) single controlled comb-shaped structure; e) double controlled comb-shaped structure; f) circle pattern.

3.2 Controlling of the Electric Field Modifier

Fig. 7 presents a simple power electronics subsystem, extended by an additional EFM control unit for the EC-MEA. The power electronics subsystem is regulating the needed DC or AC current at a specific voltage level by controlling parameters like fuel and oxidant pressures, flow rates, and relative humidities, as well as operating temperature. For this reason, a converter adjusts the load-dependent fuel cell voltage to the required level. An AC converter is needed if the fuel cell system is connected to an AC grid. To compensate transient and dynamic effects, a fast discharging electronic component like a battery or a super capacitor is used. In [7] several grid integration architectures are discussed and an optimization methodology for e.g. minimum costs, weight and volume is described.

The new EFM control unit in Fig. 7 consists of an external controllable voltage source and EFM electrodes integrated in the fuel cell. The external voltage source needs to react on a measured voltage drop. In Fig. 6, different actuating signals to control the EFM electrode are presented. Fig. 6 a) shows a constant and a pulsed DC voltage, whereas in Fig. 6 b) three different AC signals are depicted: a sinusoidal, a pulsed and a triangle voltage signal. In the case of a variable AC voltage source, different frequencies and amplitudes can be used to actuate the EFM electrode.

4 Anticipated Advantages

In the following subsections, several anticipated advantages of controlling PEM fuel cells with an EFM control unit are described and discussed.

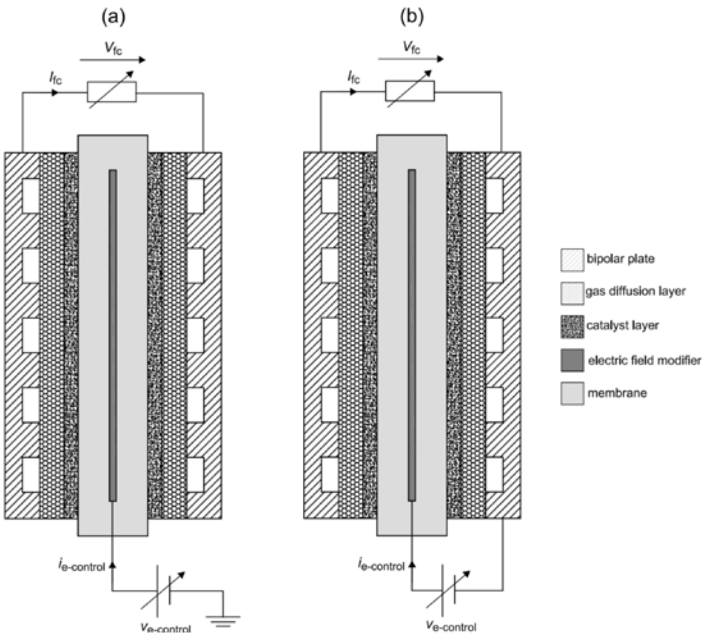

Fig. 4. Possible profiles of PEM fuel cells with a single electric field modifier (EFM) in the membrane and examples of electric circuits with a variable DC voltage source: a) The potential of the EFM is independent of the fuel cell electrode potentials; b) The potential of the EFM is dependent on the potential of the anode or cathode.

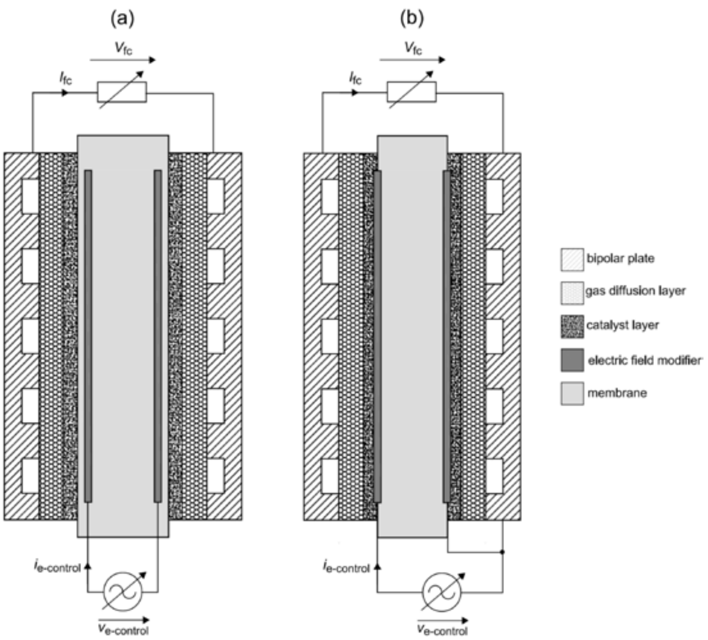

Fig. 5. Possible profiles of PEM fuel cells with two electric field modifiers (EFM) in or at the membrane and examples of electric circuits with a variable AC voltage source: a) The potential of the EFM is independent of the fuel cell electrode potentials; b) The potential of the EFM is dependent on the potential of the anode or cathode.

4.1 Transient Voltage and Power

Curve 2 in Fig. 1 b) shows a voltage behavior of actual fuel cells after a positive current step, which means a step from low to high load. Usage of EC-MEA under the same load step indicates a lower voltage drop and a faster dynamic response. This is due to the controllability of reaction kinetics in the membrane, and thus the direct effect on the proton transport through the membrane, which decreases ohmic losses for a short time. With this, steady-state

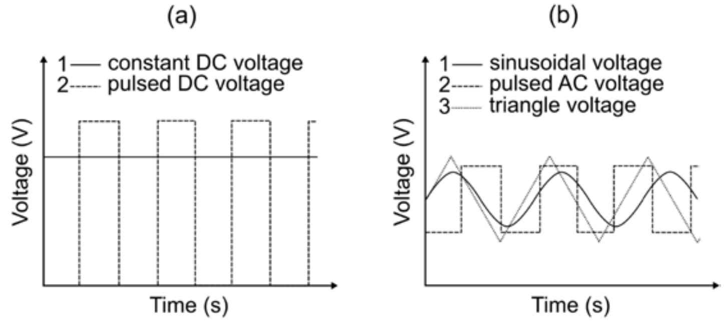

Fig. 6. DC and AC actuating signals for the electric field modifier electrode: a) Constant DC voltage (a.1) and pulsed DC voltage (a.2); b) AC actuating signals with variable frequencies and amplitudes: b.1) sinusoidal voltage; b.2) pulsed voltage; b.3) triangle voltage.

voltage and power is reached faster, and fuel cell systems with EC-MEA are able to react faster to fluctuating load conditions.

In addition to this, EC-MEA will be able to enable temporary higher current densities. This leads to temporary higher power densities, and therefore an oversizing of the fuel cell system is not needed anymore. Temporary higher power densities increase fuel cell system availability due to a late system shutdown, which is needed to protect membranes against higher temperatures.

Vayenas et al. showed similar effects of higher power densities in [16]. They modified a fuel cell by adding a third electrode in the setup to clean the CO-pollution on the anode. This led to enhanced steady-state current and power outputs, but is not suited to control the reaction kinetics over the whole membrane area and does not improve dynamic behavior.

4.2 Integration in On-Board Power Supplies

The improved dynamic behavior of fuel cell systems due to usage of an EFM control unit enables a reduction of system complexity and integration costs e.g. in on-board power supplies. As already mentioned, integration of actual fuel cell systems is connected to an system oversizing and the usage of batteries or super capacitors to compensate the slow system dynamic in the event of a load change [6], [7]. System costs C_{sys} and system weight m_{sys} can be calculated with (9) and (10):

$$C_{\mathrm{sys}} = C_{\mathrm{fc}} + C_{\mathrm{DC-DC}} + C_{\mathrm{storage}} + C_{\mathrm{DC-AC}} \quad (9)$$

$$m_{\mathrm{sys}} = m_{\mathrm{fc}} + m_{\mathrm{DC-DC}} + m_{\mathrm{storage}} + m_{\mathrm{DC-AC}} \quad (10)$$

with the costs C and weight m for the fuel cell (fc), the DC-DC converter (DC-DC), the energy storage, which can be a fast battery or a super capacitor, and the inverter (DC-AC), see Fig. 7. In both equations, the investment costs and the weight of only the main components are considered. Generally, costs and weight of the components are dependent on the power or energy of the system, as stated e.g. in [7].

The whole electrical system could be smaller dimensioned and batteries or super capacitors could be completely dropped if the system dynamic improves significantly, e. g. with the proposed EFM control unit. To decrease system costs and weight, the EFM control unit needs to cost and weigh less than the achievable savings of the whole electrical system due to sufficient dimensioning. With this, the following simple inequalities (11) and (12) determine the development objectives:

$$C_{\mathrm{fc,new}} + C_{\mathrm{DC-DC,new}} + C_{\mathrm{storage,new}} + C_{\mathrm{DC-AC}} \leq C_{\mathrm{sys}} \quad (11)$$

$$m_{\mathrm{fc,new}} + m_{\mathrm{DC-DC,new}} + m_{\mathrm{storage,new}} + m_{\mathrm{DC-AC}} \leq m_{\mathrm{sys}} \quad (12)$$

with the investment costs C and weight m of the new dimensioned main components due to the improved fuel cell dynamic. Costs and weight of the EFM control unit are included in $C_{\mathrm{fc,new}}$ and $m_{\mathrm{fc,new}}$. The inverter costs and weight is not changed, because the same total power needs to be delivered.

Furthermore, the control concept with an EFM control unit and the related temporary increase of current density enables an expected provision of higher short-circuit currents. This allows for the usage of simple state of the art grid protection concepts, which are not usable today because of too low short-circuits. In comparison, actual protection concepts of fuel cell systems are more complex and costly.

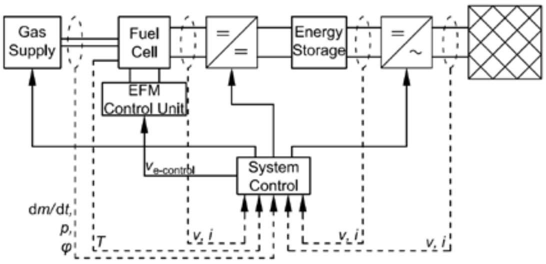

Fig. 7. New simple power electronics subsystem with an electric field modifier (EFM) control unit, own representation and extension according to [17]: reactant flow rate $\mathrm{d}m/\mathrm{d}t$, reactant partial pressures p, reactant relative humidity φ, operating temperature T, fuel cell voltage v, fuel cell current i, and internal controlling voltage $v_{\mathrm{e-control}}$.

4.3 Membrane Durability

Besides the targeted improvement of dynamic behavior of PEM fuel cell systems, usage of an EFM control unit aims to regulate and homogenize current density and temperature distribution across the membrane. With this, temperature hot spots can be avoided and membrane durability increases [9]. For this, it is essentially to use an EFM electrode setup with multiple electrodes, which can be controlled individually. The EFM control unit then needs to respond to a measured current density in the fuel cell.

4.4 Future Work

Future work consists of simulative and experimental investigations of the optimal design, implementation and connection of EFM electrodes in the fuel cell setup. Main objective here is a maximum three-phase-boundary of the EC-MEA while enabling an optimal control of the dynamic behavior. In addition, ideal electrode material, dimensions, and structures need to be identified. Finally, an optimal way of actuating the EFM electrodes with the EFM voltage source needs to be analyzed.

5 Conclusion

Fast changing loads are still a challenge for current PEM fuel cell systems due to the relatively slow dynamic behavior. Present control strategies involve regulation of operational parameters like flow rates and pressures, which result in unstable fuel cell voltage in the order of seconds to minutes. Consequently, fuel cell systems are oversized and costly storage systems for peak power, transient voltage, and power compensation are integrated to compensate for slow system dynamics. This contribution presents a new way of controlling PEM fuel cells by means of internal effects using an electrically controllable membrane electrode assembly (EC-MEA). An appropriate design and integration of electric field modifier (EFM) electrodes into the membrane electrode assembly is important to take full advantage of the discussed benefits. The expected benefits of EC-MEA e.g. an improved transient voltage behavior and a greater power density result due to preliminary considerations based on proton transport mechanisms of PEM fuel cells.

Next steps include the development and experimental characterization of different EFM electrode structures and EC-MEA configurations. Moreover, an optimal actuating concept of the EFM electrodes to control dynamic voltage behavior is going to be investigated.

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