Using of an Electrochemical Compressor for Hydrogen Recirculation in Fuel Cell Vehicles

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Received May 09, 2019; accepted May 19, 2020; published online June 08, 2020

Abstract

The automotive industry sees hydrogen-powered fuel cell (FC) drives as a promising option with a high range and short refueling time. Current research aims to increase the profitability of the fuel cell system by reducing hydrogen consumption. This study suggests the use of an electrochemical hydrogen compressor (EHC) for hydrogen recirculation. Compared to mechanical compressors, the EHC is very efficient due to the almost isothermal conditions and due to its modular structure, can only take up a minimal amount of space in vehicles. In addition, gas separation and purification of the hydrogen takes place in an EHC, which is a significant advantage over the standard recirculation with a blower or a jet pump. The high purity of the hydrogen at the cathode outlet of the EHC, also increased partial pressure of the hydrogen at the fuel cell inlet and its efficiency. The study carried out shows that replacing the blower with the EHC reduces the hydrogen loss by purging by up to ~95% and the efficiency of the FC system could be further improved. Thus, the EHC has a great potential for recycling hydrogen in FC systems in the automotive industry and is a great alternative to the current blower.

Keywords: Electric Power Consumption, Electrochemical Hydrogen Compressor, Fuel Cell, Hydrogen Efficiency, Hydrogen Pump, Hydrogen Purification, Hydrogen Recirculation

1 Introduction

1.1 Short Description of the Necessity for Recirculation

The most common fuel cell used in vehicles is the polymer electrolyte membrane fuel cell (PEMFC). The PEMFC runs on hydrogen and air. Adequate supply with reaction gases is critical for high performance and the lifetime of the PEMFC.

Typically, more hydrogen and air are pumped through the fuel cell than is needed for the electrochemical reaction. Therefore, a fuel starvation and an air starvation can be avoided, this increases the life of the fuel cell [3].

Hence, the hydrogen ratio $\lambda_{H_2}$ and the air ratio $\lambda_{Air}$ are usually greater than 1. The hydrogen ratio describes the ratio of the hydrogen inlet mole flow $n_{H_2, FC,in}$ to the hydrogen mole flow, needed by the electrochemical reaction $n_{H_2, FC,use}$.

\[ \lambda_{H_2} = \frac{n_{H_2, FC,in}}{n_{H_2, FC,use}} \] (1)

The air ratio is similar and is the ratio of the supplied air mole flow $n_{Air, FC,in}$ to the required air mole flow $n_{Air, FC,use}$.

\[ \lambda_{Air} = \frac{n_{Air, FC,in}}{n_{Air, FC,use}} \] (2)

Thus, the excess hydrogen at the anode exhaust gas is pumped back to the anode inlet of the FC stack with the help of an active blower or a passive jet pump or both and combined with fresh hydrogen from the tank (Figure 1) [3].
Due to the diffusion and permeation through the membrane, gas exchange takes place.

This transport process generally works in both directions, but the gases preferably migrate in the direction of the lower concentration or the lower partial pressure (Figure 2) [1].

The parasitic gas exchange of hydrogen and oxygen leads to the production of water vapor and causes additional humidification of the membrane electrode assembly (MEA). Operating with air has the disadvantage that the nitrogen contained in the air also diffuses to the anode side. Due to the recirculation in the anode circuit, this leads to a concentration of inert gases. Therefore, on the anode side of the EHC, the electrochemical compressor is a modified polymer electrolyte membrane fuel cell, which is connected to a constant current source for pumping and separating hydrogen from a hydrogen containing gas mixture [3, 4]. Thus, there is no electrochemical energy conversion as in a fuel cell, but an increase in pressure without mechanically moving components [1, 2, 9–11].

1.2 Replacing the blower with an Electrochemical Compressor

This study suggests a new option to recycle unused hydrogen in a fuel cell system, using an electrochemical compressor (EHC) instead of a blower (Figure 3).

The electrochemical compressor is a modified polymer electrolyte membrane fuel cell, which is connected to a constant current source for pumping and separating hydrogen from a hydrogen containing gas mixture [3, 4]. Thus, there is no electrochemical energy conversion as in a fuel cell, but an increase in pressure without mechanically moving components [1, 2, 9–11].

1.3 Function of an Electrochemical Compressor

Figure 4 shows that the structure of the electrochemical compressor is very similar to the structure of a PEMFC in Figure 2. In contrast to the fuel cell, however, no gas is supplied to the electrochemical compressor on the cathode side from outside. Thus, no reaction between hydrogen and oxygen is possible here. At the anode side of the EHC, the hydrogen is oxidized. The protons are conducted through the membrane by ion conduction, while the electrons flow to the switched-on constant current source. The constant current source creates an electrical potential between the electrodes. At the cathode side, the electrons and protons recombine to hydrogen, and continuous pumping increases the pressure on the cathode side in a closed system. Figure 4 also shows that the EHC separates gases, because only hydrogen protons are pumped through the membrane. Other gases such as nitrogen are unaffected by the pumping action, stay on the anode side and can be removed. Only by diffusion processes, other gases are transported from the anode to the cathode side of the EHC [1, 2, 4, 5].

In addition, the EHC is robust and requires little maintenance as it contains no moving parts. In addition, there is quiet operation and the possibility of a modular structure. By using components similar to those used in the FC, there is potential for integration into a FC stack. Furthermore, due to its almost isothermal operation, the EHC is more efficient than mechanical compressors. The main tasks of the electrochemical compressor in the proposed application could thus be the separation of hydrogen from the fuel cell exhaust gas mixture and the pumping of the extracted hydrogen [9–11].
2 Experimental and Numerical Methods

2.1 Simulation

Within the scope of this work simulations were performed with the simulation software ComsolMultiphysics Version 5.1. In addition to the basic module of ComsolMultiphysics, the module “Batteries and Fuel Cells” was used for the simulation.

Since the anode exit of the FC stack supplies the entry parameters for the EHC, Figure 3, a simulation of a reference FC stack was first carried out. The operating conditions of the reference FC stack for vehicle use were defined, in order to characterize the time-dependent transport processes of the gases.

The most important parameters for the simulation are listed below [1, 5–7].

(i) The reference is made to the current/voltage characteristic of a FC stack with an electrical output of 100 kW. The FC stack voltage is taken from the current/voltage characteristic curve (Figure 5) according to [6].

(ii) The FC stack has 370 MEAs, each with an active area of 320 cm².

(iii) The dry membrane thickness $t_{\text{Mem}}$ is 25.4 $\mu$m.

(iv) The starting process is idealized in the absence of nitrogen.

(v) The internal transport processes of the gases in the fuel cell are considered separately and quantified as contamination of the hydrogen in the anode circuit.

In practice, the required gas ratio in FC vehicles is mostly achieved by a combination of jet pump and recirculation blower, as shown in Figure 6.

The gas composition and the thermodynamic values of the anode exhaust gas from the FC stack were determined and used for the simulation and design of the EHC in the vehicle. The EHC is mainly used in the low-load area of the FC stack (Figure 6). The jet pump supports the EHC with increasing load and takes over the complete recirculation in the high-load range.

Similar geometrical assumptions were used to simulate the EHC. The dimensions of the EHC-MEA were selected similar to the FC-MEA, i.e.,

(i) the dry membrane thickness $t_{\text{Mem}}$ is 25.4 $\mu$m;

(ii) and the active area is 320 cm².

The anode inlet of the EHC is therefore connected to the anode outlet of the FC, so that a gas mixture of hydrogen, water vapor and nitrogen is present on the EHC anode. The pressure difference between the anode and cathode side of the EHC corresponds to the pressure loss in the FC stack on the anode side. The EHC is isothermal and the operating temperature should initially be 80°C in all operating points.

The EHC-MEA is moistened by absorbing the water from the anode side of the FC stack. The relative humidity of the EHC cathode is set by the water transport mechanism by the MEA. Here, the internal transport processes of the gases in the EHC are considered separately, too.

2.2 Experimental

It was experimentally tested whether the theoretical relationships between the current density and resulting volume follow the Faraday law without any restrictions.

A Umicore pMembrain H300 reference cell with an active area of 100 cm² and a membrane thickness of 25 $\mu$m were used for the experiments. Carbon electrodes with a platinum loading of 0.4 mg cm⁻²
were attached directly on each side of the membrane as anode and cathode. A Sigracet GDL 10 BB gas diffusion layer (GDL) with a somewhat larger dimension of 103 cm$^2$ was also applied directly to the electrodes. The flow field for the hydrogen and oxygen side consists of two laser-welded stainless steel sheets, each with an 8-fold meander. The space between the welded sheets was used to transport demineralized water to cool the cell. Sealing frames are used as sealing elements, which consist of a stainless steel frame and screen-borne sealant. The stainless steel current collectors are built into the epoxy resin end plates. Connections for connecting the media lines are screwed into the end plates. The components of the electrochemical compressor are pressed with a contact pressure of 1.5 N mm$^{-2}$.

Several series of measurements were carried out with pure hydrogen and a gas mixture of 60% hydrogen and 40% nitrogen at different temperatures and relative humidity. The tests with pure hydrogen have been used as a reference measurement for the electrochemical compressor and to investigate the effect of temperature and humidity.

A gas mixture of 60% H$_2$ and 40% N$_2$ was humidified and fed to the anode inlet of the EHC after adjusting the flow rate. The pressures, temperatures and the H$_2$ mole fraction were measured. Several experiments were carried out at different temperatures (50–90°C) and relative humidity (60–90% r.H.). The operating pressure was 1.5 bar on the EHC anode side and 1.6 bar on the EHC cathode side. So that the pressure difference between the anode and cathode side of the EHC almost corresponds to the pressure loss in the FC stack on the anode side.

The EHC was operated at a constant flow rate and the voltage-current characteristics were recorded. The relative humidity, flow rate, temperature, and H$_2$ mole fraction of hydrogen were determined on the cathode side. In the next step, the series of measurements were repeated with higher temperatures and/or air humidity.

The flow rates from the experiment were then compared with the theoretical flow rates according to Faraday’s law in direct dependence on the current and with reference cells from the literature.

### 3 Results and Discussion

#### 3.1 Inert Gas Discharge

In the vehicle system, nitrogen contents of 2–5% are realistic on the anode outlet of the fuel cell. Therefore, at first, a simulation with a constant gas composition with 98% H$_2$ and 2% N$_2$ at the EHC anode inlet was examined. A pressure difference between the anode and the cathode side of the EHC was set to 100 mbar in all experimental investigations. This corresponds to the pressure loss on the anode side of an FC stack in a vehicle [3].

During the pumping process in the simulation the current density of the EHC was 1.13 A cm$^{-2}$, the hydrogen was pumped to the cathode side, whereby the hydrogen mole fraction at the anode side along the channel was reduced from $\sim$96% to $\sim$2.5% (Figure 7). The total volume flow decreased and there is an increase of the nitrogen mole fraction from 4% to $\sim$40% along the length of the cell (Figure 8), whereas the amount of substance of nitrogen stays constant.

The nitrogen diffusion and permeation from the anode to the cathode also takes place in the EHC, but in a much smaller amount than in an FC since the partial pressures are lower here. Thus, the N$_2$ mole fraction in Figure 8 is shown as a percentage on the anode, while the cathode side is in ppm. The highest mole fraction of $\sim$80 ppm nitrogen was found in the dead end area of the cathode. At the cathode outlet of the EHC, the nitrogen content mixes with the pumped hydrogen and the concentration drops to $\sim$40 ppm. So, one will find almost pure, wet hydrogen at the cathode outlet of the EHC [1, 10, 11].
Similar purification effects could be determined in the simulations of FC and EHC in series with performance dependent gas composition.

For the experiment a hydrogen/nitrogen mixture with a gas composition of 60% hydrogen and 40% nitrogen at the anode inlet of the EHC has been used. The experiment showed that almost all of the nitrogen could be separated from the hydrogen and that there was only a small amount of nitrogen on the cathode side of the EHC. From the original gas composition with 60% hydrogen and 40% nitrogen at the anode inlet of the EHC, there are now around 99.4% hydrogen and only 0.6% nitrogen at the cathode (on a dry basis, Figure 9). The cleaning effect by the EHC was confirmed in the experiments.

To revise the simulation, a gas composition of 98% hydrogen and 2% nitrogen at the anode inlet was also tested. No nitrogen content in the cathode gas could be found with the built-in sensor. Examination of the gas with a gas chromatograph showed a nitrogen content of ~40 ppm at the cathode of the EHC.

### 3.2 Water Balance

As in the case of a FC, the electro-osmosis, diffusion and permeation of an EHC transports water molecules from the anode to the cathode [13, 14].

The relative humidity at the cathode is influenced by two factors. The main part is the supply of humidified hydrogen to the EHC anode, the more humidity the supplied gas has, the more water is transported to the cathode side. The second mechanism for increasing the relative humidity from the anode to the cathode side is due to the pressure increase. Since the relative humidity represents the ratio of the water partial pressure to the vapor pressure, the relative humidity increases with the same amount of substance as the pressure increases.

By varying the relative humidity and temperature at the EHC anode inlet, the optimum inlet relative humidity can be determined. The humidification is designed so that the cathode outlet of the EHC has the highest possible relative humidity, but at any point of operation of the EHC, the relative humidity never exceeds the saturation limit (Figure 11).

In the experimental studies, the relative humidity was gradually increased and the effects on the current-voltage character were observed. Results of this experiments are shown in Figure 11. The lower the voltage is with constant current density, the lower is the power consumption with constant pumped mole flow. The lower the relative humidity is (below 80% r.H.) and the lower the temperatures are (up to 80 °C) the current-voltage characteristics become worse.

The best results were achieved with a relative humidity of 80–82% on the anode side. The cathode gas at the outlet had reached a maximum relative humidity of 97–99% (Figure 12).
At higher relative humidities, the gas on the cathode side exceeds saturation and water condenses. This effect worsens the current-voltage characteristic.

### 3.3 Electric Power Consumption

Figure 13 shows the recorded U/I characteristic of a 60% H₂ gas mixture at the temperature of 80°C and a relative humidity of 80%. It is compared with the U/I characteristic of the pure hydrogen with the same humidification and temperature. It should be noted that the flatter the U/I characteristic, the better the efficiency. Due to the lower limit of the mass flow controllers (MFCs), the range of the activating voltage at low currents is not shown.

EHC studies with a hydrogen-gas mixture or pure hydrogen show increasing mass transport limitations at higher flows. Mass transport limitations due to decreasing mole fraction of hydrogen hinder the transport of the gas through the GDL to the anode. As a result, there is an increase in the voltage and energy consumption and thus, the efficiency of the hydrogen pump is lowered [19].

#### Table 1: Vehicle model parameters.

| Vehicle model parameters | Symbol | Values | Units |
|--------------------------|--------|--------|-------|
| Vehicle total mass       | m      | 1,300  | kg    |
| Aerodynamic drag coefficient | c_w  | 0.25   |       |
| Frontal area             | A      | 2      | m²    |
| Tire drag coefficient    | c_r    | 0.012  |       |
| Tire size                |        | 215/55 R17 |       |
| Effective rolling radius of wheel | 0.345  | m      |       |
| Auxiliaries consumption  | 1      | kW     |       |
| Walk & slip efficiency   | 95%    |        |       |
| Transmission efficiency  | 95%    |        |       |
| Engine efficiency        | 90%    |        |       |
| Converter efficiency     | 95%    |        |       |
| Battery efficiency       | 90%    |        |       |
when the nitrogen content reached 5% and closed when it dropped below 2%.

The simulation results were used to determine a physical efficiency chain and power losses of individual components as well as the hydrogen consumption of the entire system.

In this calculation in WLTP, the energy consumption of the circulation pump was 130 Wh and the purge loss was 559 Wh, which corresponds to 7% of the hydrogen from the tank.

The recirculation with an EHC and jet pump was simulated in a similar way. The simulation shows that recirculation with an EHC leads to a loss of hydrogen by purging of 27 Wh and the energy consumption of the EHC is ~83 Wh.

It was found that the hydrogen consumption for the EHC was lower than that of the recirculation blower, the overall losses of the FC system could be reduced and the efficiency from tank to wheel could increase from 37% to 39% in the WLTP (Table 2).

3.4 Mechanical Integration of EHC

Two options are proposed for the mechanical integration of the EHC into the FC system [2].

Alternative A (Figure 15) is the integration of the EHC in series with the FC stack behind the FC stack and Alternative B (Figure 16) is the integration of the EHC in series with the FC stack in front of the FC stack.

Similar MEAs and bipolar plates could be used in both cases and offer the best use of space in the vehicle. By integrating the EHC in line, the existing FC stack is 6 to 10 cm longer, but this saves additional components such as hoses. The circulation takes place in a small circuit nearby or directly in the FC-EHC-combi-stack.

An additional advantage of alternative B is that fresh hydrogen from the tank flows through the cathode side of the EHC. So there is a mixing between pumped and fresh hydrogen.

As described in Section 3.2, water is transferred from the anode to the cathode. This process is limited by the hydrogen saturation on the cathode side of the EHC. Since the cathode now is flowed through and the relative humidity is thereby reduced, more water can now be transported from the anode to the cathode of the EHC (Figure 17).

This effect could be further enhanced by increasing the temperature of the EHC by 5–10 K. Since the relative humidity represents the ratio of the water partial pressure to the vapour pressure, the relative humidity rises proportionally to the vapour pressure.

In both cases, the modular structure of the EHC results in better weight and space utilization of the FC system. By using alternative B, a greater relative humidity can be achieved at the outlet of the EHC. This can result in better FC stack humidification on the anode side, which allows a reduction or elimi-
ination of the FC cathode humidifier. In addition, the EHC is robust, works very quietly and requires little maintenance as it contains no moving parts.

4 Conclusions

The simulations and experiments carried out in this work show that the EHC has a good pumping and cleaning effect and the efficiency of the FC system can be improved by using electrochemical compressors. Especially the cleaning effect could be very interesting for the future if impurities from the tank are found in the FC system.

By replacing the blower with the EHC, the hydrogen loss from the purge in the WLTP can be reduced from 559 Wh to 27 Wh. This means that the hydrogen loss in the purge can be reduced by up to ~95%. The use of an EHC further improved the efficiency from tank to wheel by ~2%. Hydrogen consumption with the EHC is lower and the efficiency of an FC vehicle could be increased from 37% to 39%.

In the simulation, a pressure increase from the anode to the cathode side of 0.1 bar and a hydrogen recovery of 98% with a possible hydrogen mole fraction (dry basis) of over 99.9% were realized. The simulation of the EHC in the WLTP shows a similar power consumption like the blower.

Due to the large recovery of hydrogen, the EHC has a considerable advantage over the recirculation blower. Due to the recirculation, the purity of the hydrogen at the cathode outlet of the EHC also leads to an increasing partial pressure of the hydrogen at the fuel cell and thus increases its efficiency. This leads to a better overall efficiency of the FC system.

The EHC has no moving parts and therefore requires little maintenance, works quietly and is very robust. Production costs can be saved by using the same components as in the FC, such as flow fields and gas diffusion layers. In addition, the FC series design can reduce the weight and volume of the FC system, further increasing efficiency.

Thus, the electrochemical compressor has great potential for use in the circulation in the automotive industry and is a good alternative to today’s circulation blowers. The EHC could play an important role in the development of fuel cell vehicles and help reduce hydrogen consumption, which will lead to more energy-efficient and environmentally friendly mobility in the future.

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