



Towards re-electrification of hydrogen obtained from the power-to-gas process by highly efficient H₂/O₂ polymer electrolyte fuel cells

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ARTICLE

Towards re-electrification of hydrogen obtained from the power-to-gas process by highly efficient H₂/O₂ polymer electrolyte fuel cells

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In the power-to-gas process, hydrogen, produced by water electrolysis is used as storage for excess, fluctuating renewable electric power. Reconversion of hydrogen back to electricity with the maximum possible efficiency is one pre-requisite to render hydrogen storage technically and economically viable. Pure oxygen is a byproduct in the electrolysis of water. The use of pure oxygen as the oxidant in a polymer electrolyte fuel cell (PEFC) is a possible way of increasing the conversion efficiency of hydrogen to power, by reducing the fuel cell's cathodic kinetic overvoltage, which is the most important energy loss process in low temperature PEFCs. As we demonstrate in this work, when using pure oxygen, either high efficiencies at current densities around 1 A/cm² are obtained or a very high power density operation (up to 1.6 W/cm² at cell voltages above 0.62 V) can be reached, giving the technology a broad window of operation and application. The fuel cell stack durability is assessed in accelerated long-term tests of up to 2'700 h. The potential of the technology is demonstrated with the realization of a complete 25 kW prototype system delivering a peak efficiency of 69 % LHV (57% HHV).

Broader Context

Not only, but most importantly because of the concern about global warming, capacities for generation of renewable power are being increasingly deployed. As the temporal pattern of production for many of the renewable power sources cannot be controlled, and in the absence of storage capacity of the grid, the historical paradigm that production needs to match demand in the electric grid may gradually change to demand to match production. This requires means of storage for electric power on a diurnal and maybe even seasonal scale. Therefore different technologies for storage of electric energy are being developed. One of the investigated power storage technologies is the so-called power-to-gas concept, where excess power is locally used to split water using the water electrolysis process. Hydrogen and oxygen produced can be used along different pathways, i.e. to produce synthetic natural gas, in hydrogen based mobility (fuel cell cars) or can be reconverted to power in order to satisfy demand in periods of low production. In the economic and technological context efficiency is a key attribute of all the conversion technologies employed.

1 Introduction

In a future sustainable energy system, based on a large fractional contribution from renewable power sources (mainly solar and wind technologies), energy storage will be essential to balance production and consumption within the electric grid. Energy storage itself is crucial for renewable energy sources in order to avoid curtailment.¹

Curtailment is a global issue², in Germany, today's largest producer of wind and solar power in Europe, approximately 100 GWh of wind power remained unused in 2010.³ This number is expected to grow to 15 TWh in 2030 without suitable energy storage solutions.⁴

Among the energy storage technologies under investigation, power-to-gas^{5,6} i.e. electrolyzing water to produce hydrogen and/or synthetic natural gas, promises to provide solutions on several different levels. These include large scale seasonal energy storage in caverns or the natural gas grid (in the TWh-range) to distributed grid stabilization units (in the GWh-range). If re-electrification of the gas is in the focus of the power-to-gas process, the direct conversion of hydrogen into electrical energy potentially offers significantly higher round-

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trip efficiencies as compared to its further conversion into synthetic natural gas followed by combustion in a gas turbine. Furthermore, hydrogen obtained from this process may also play an important role as a fuel in future sustainable road transportation.

The difficulty in achieving high electron-to-electron round-trip efficiencies is considered the main drawback of power-to-gas energy storage. Using low temperature electrolyzers (alkaline or polymer electrolyte, with efficiencies of 70 to 80% (HHV) and conventional H₂/air fuel cell technologies (typically < 50% HHV), round-trip efficiencies below 40% are reached.

However, when considering the best technologies for both processes, for electrolysis, the solid oxide high temperature technology, providing up to 90% (HHV) efficiency⁷ and optimized H₂/oxygen fuel cells (up to 60% HHV), round-trip efficiencies above 50% become feasible.

When hydrogen is produced by water electrolysis, pure oxygen is obtained as an energetically inexpensive by-product. In the power-to-gas technology, utilization of this by-product may result in a strategic efficiency advantage.

The common oxidant used in polymer electrolyte fuel cell systems (PEFC) is oxygen from the air. However, the most significant energy loss mechanism in PEFCs originates from the oxygen reduction reaction (ORR), inherently related to the oxygen partial pressure at the cathode. The use of pure oxygen can increase the partial pressure of oxygen by an order of magnitude, thus significantly reducing the activation losses (i.e. high overpotentials) due to the sluggish ORR.

In addition to reducing cathodic activation overpotentials, using pure oxygen over air as the oxidant in a PEFC offers several further advantages, which include:

- i) Higher specific power (mainly a significant cost advantage, as a stack of only about half the size is required to obtain the same power).
- ii) Higher system efficiency (even if operated at the same cell voltage as the air system, the absence of air compression in the oxygen system means that the balance of plant has a lower consumption).
- iii) Higher power and start-up dynamics (no limitations by the “slow” air path).
- iv) Easier water management (no nitrogen to be humidified).
- v) Less dependence on environmental conditions (altitude, humidity, pollution by process air).

The use of elevated oxygen partial pressures however also raises a number of questions. There might be durability issues, which could shorten the service life, in particular chemical degradation processes related to the oxygen partial pressure.

There is fundamental consideration in the literature^{8,9} concerning the effect of the oxygen partial pressure in PEFCs, but little information regarding the specific performance, electrochemical cell and system efficiencies and durability with pure oxygen as the cathode oxidant is available.¹⁰⁻¹⁴ Submarines have so far been the only industrial application for

PEFC H₂/O₂ technology. For space applications NASA has used alkaline systems since the 1960s.

In order to assess the feasibility of hydrogen re-electrification in the power to gas energy chain with H₂/O₂ PEFC technology, in this paper we explore the specific power density achievable in cells and short stacks, analyse the effect of the elevated oxygen partial pressure on efficiency and durability, and finally compare the results of a proof of concept system with a commercial hydrogen/air system of same stack size and similar rated power.

2 Experimental

All experiments were performed in full size hardware with an active area of 238 cm². The cell has a *circular* design with cascaded flow fields for anode and cathode to sustain a quasi-constant gas velocity for the pure reactants over the active area.¹⁵ Bipolar plates were made from graphite composite material, the cooling flow field is integrated in the middle of the two part plates and for sealing of gases and coolant molded olefinic gaskets are used.

As the electrochemical components, commercial membrane electrode assemblies (MEAs) of the type H400E (Solvicore, D) based on Dupont's Nafion XL100 membrane (a dry thickness of approx. 30 μm) with catalyst loadings of 0.1/0.4 mg/cm² (A/C) were used.

If not mentioned otherwise cells and stacks were operated using a set of standard conditions, as given in Table 1 for experiments in flow through mode on test benches. In system configuration, gas stoichiometries and gas dew points were not controlled/measured.

Current density distributions were measured with a Current ScanLin Device from S++ (Munich, D) with 440 individual segments of a size of 7 x 7 mm. Adjoining bipolar plates were designed specifically to retain thermal and fluid dynamic properties as similar as possible as for “normal” cells in a stack.

For durability experiments cyclic current profiles were applied as accelerated stress tests as shown in Figure 1. The given current densities translate into absolute currents of 40-80-200 A for the low and 80-160-400 A for the high current profile. The two profiles include a common current level of 80 A (0.34 A/cm²) so durability can be compared between the two profiles.

Table 1: Base case operating conditions for sub-stack tests.

Property	Value
Cell Temperature [°C]	74
Gas pressure A/C [MPa]	0.25 / 0.25
Gas inlet dewpoint A/C [°C]	60 / 60
Gas inlet rel. humidity [-]	0.54 / 0.54
Gas stoichiometry A/C [-]	1.3 / 1.5

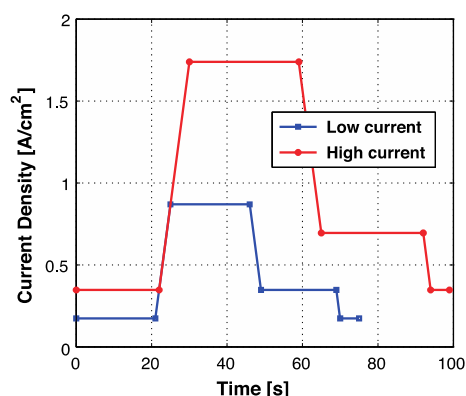


Figure 1: Current profiles used for durability experiments; translate into absolute currents of 40-80-200 A for the standard profile and 80-160-400 A for the high current profile.

In order to estimate the sources of degradation in long-term experiments, a simple model was applied:

$$E_{\text{cell}} = E_{\text{OCV}} - b \cdot \log\left(\frac{i}{i_0}\right) - R \cdot i \quad (1)$$

where R is the ohmic area resistance, b the Tafel-slope, E_{OCV} the apparent open circuit voltage, i_0 a constant representing the exchange current density and i the current density. The three different load points in the durability experiments represent a simplified load curve, so the parameters in eq. (1) can be fitted and followed over time.

Efficiency considerations

For fuel cells, commonly the efficiency is calculated with respect to the lower heating value of hydrogen (LHV), while in the electrolysis community for determining efficiency the higher heating value (HHV) is used. In both cases better values are calculated for the individual technologies. In this work we report measured efficiencies for the fuel cell technologies also with respect to LHV in order to make them comparable with previous literature. However when calculating round-trip efficiencies power-to-hydrogen (and oxygen) to-power then for both conversion steps the same basis needs to be considered. So when reporting round-trip efficiencies, we relate the fuel cell values also to HHV, in order to make correct estimates.

3 Results and Discussion

Performance

Oxygen reduction is the main loss mechanism in acidic low temperature polymer electrolyte fuel cells. The Tafel equation relates the kinetic loss, termed overvoltage η to the exchange current density i_0

$$\eta = b \cdot \log\left(\frac{i}{i_0}\right) \quad (2)$$

with the Tafel slope b being a constant with a theoretical value of $2.3RT/F$ (69 mV under the current conditions) and i the

geometric current density. The apparent exchange current density i_0 depends on the electrode roughness factor f_r (i.e. the product of catalyst loading in g/cm^2 (geometric) and the catalyst specific surface area in $\text{cm}^2(\text{specific})/\text{g}$) the specific exchange current density $i_{0,S}$ (in terms of A/cm^2 of catalyst surface) and the oxygen partial pressure p_{O_2} as

$$i_0 = 10 \cdot f_r \cdot i_{0,S} \cdot \left(\frac{p_{\text{O}_2}}{p_{\text{O}_2}^{\text{ref}}}\right)^\gamma \quad (3)$$

The exponent γ (the kinetic reaction order with respect to the oxygen partial pressure) is not precisely known, but measurements indicate a range of $\gamma \approx 0.5 - 1$ (see i.e. ^{16,17}). For reducing the cathodic overvoltage η , the exchange current density i_0 needs to be increased. As deduced from equation (3) three basically different routes are available:

- (i) increasing the roughness factor f_r by increasing loading or decreasing particle size: load-increase is limited by cost and mass transport limitations in the catalyst layer, reducing particle size by the catalyst stability. Both are therefore limited, if not undesired options;
- (ii) increasing the catalysts activity (i.e. raise $i_{0,S}$ in eq. (3)). This is the main path pursued and thus a broad research field.¹⁸ In the past 20 years, the development of alloy catalysts has led to an increase of $i_{0,S}$ by about a factor of 2 to 5.¹⁹⁻²¹
- (iii) increasing the oxygen partial pressure. This is easiest when using pure oxygen. This increase is typically about an order of magnitude (0.03 to 0.3 MPa), which is equivalent to an increase of the apparent exchange current density i_0 by a factor of three to ten, depending on the value of γ .

Apart from the kinetic improvement, the use of pure oxygen also reduces transport overvoltage under high current density conditions. The slow inter-diffusion of diluted oxygen in nitrogen when using air in the porous structures of the gas diffusion and catalytic layers is eliminated. Also the ohmic losses decrease because the membrane is kept easier in a humid, well conducting state without the high volume flow of nitrogen at the cathode. Thus, with the use of pure oxygen at the cathode all relevant losses are reduced and thus considerably higher efficiencies as compared to the operation of PEFC with air as the oxidant are achieved. A current/voltage characteristic is shown in Figure 2 for a gas pressure of 0.35 MPa. At 0.7 V the current density is about a factor of 2 higher as compared to hydrogen/air operation with the same electrochemical components under conditions (gas humidification, gas pressure) which can reasonably be realized in PEFC systems (see again Figure 2).

There are also efficiency advantages on the system level for the pure oxygen system over hydrogen/air technology, because also the cathode reactant is simply expanded from a pressure reservoir. In the absence of a blower/compressor unit for the cathode gas loop, the parasitic system power is less.

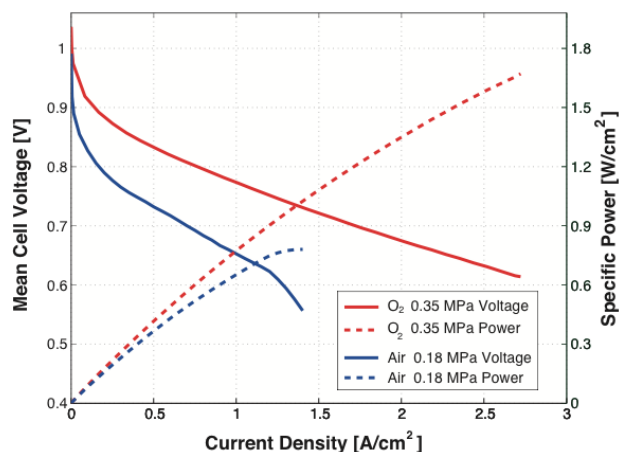


Figure 2: Current/voltage characteristics of 238 cm² cells at 74 °C cell temperature. H₂/O₂ gas pressures of 0.35 MPa, and stoichiometries anode/cathode of 1.3/1.5. H₂/air gas pressures 0.18 MPa and stoichiometries of 1.3/1.8. Data obtained on 3-cell substacks.

The results in Figure 2 show that the operation with pure oxygen basically allows for two different operation strategies: i) at cell voltages around 0.8 V, offering electrochemical cell (and system) efficiencies above 0.65 (LHV) at current densities comparable to hydrogen/air systems or ii) at current densities above 2 A/cm², with similar cell voltages/efficiencies as for H₂/air cells.

With hydrogen/air operation the current density normally has a reasonably uneven distribution along the air flow path. Differences of more than a factor of two in local current density are not uncommon under realistic operating conditions.²² The significant imbalance is due to under-humidified gas feeds and a considerable change of the oxygen partial pressure along the cathode gas stream. This current density imbalance over the active area is contributing to an increased total overvoltage of the cell. A homogeneous current density distribution allows for the maximum cell voltage with the given electrochemical components.

In the case of operation with pure oxygen, except for the pressure drop in the flow field and the changing water vapour pressure, the oxygen partial pressure (p_{O_2}) is constant. At the condition of 0.25 MPa outlet pressure, the variation in p_{O_2} does not exceed 15% except for high cell temperatures and very low feed gas humidity (i.e. the 95 °C / rH=0.2 case in Figure 3). In fact Figure 3 shows that only this hot and dry case shows a severely uneven current density distribution with low current density near the gas inlets at the top of the cell. The standard deviation of the current density of all 440 measured segments is $\sigma_j = 0.44$ A/cm² at the average current density of 1.25 A/cm². Already at a cell temperature of 75 °C with gas-inlet humidities of rH=0.2, the standard deviation drops to $\sigma_j = 0.17$ A/cm² at the same total current. For the apparently homogeneous current density distribution at 55 °C a standard deviation of $\sigma_j = 0.10$ A/cm² at 1.25 A/cm² is calculated. The slightly higher

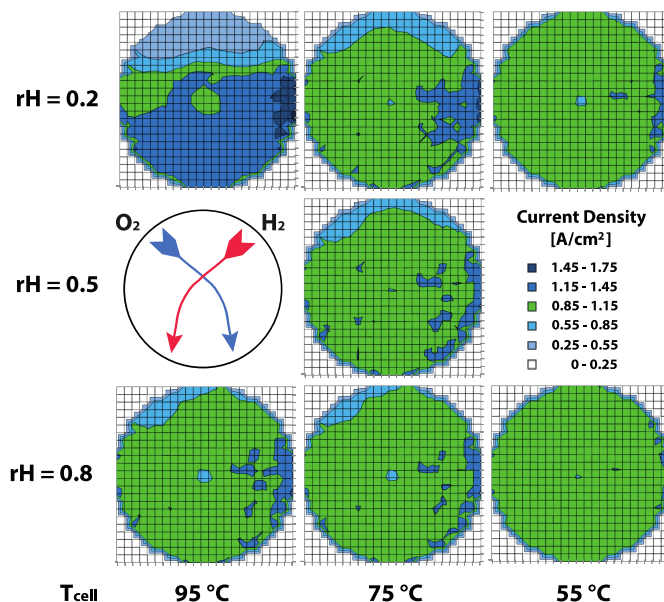


Figure 3: Current density distribution at 1.25 A/cm² (average) with $p_{gas} = 0.25$ MPa and gas stoichiometries of 1.3/1.5 (A/C) for the temperatures (columns) and relative gas humidities (rows) indicated; gas flow geometry indicated in graph middle/left; data obtained on single cell.

current density at the right edge of the cell is caused by the location of the tap of the current collector, an artefact of the single cell measurement at the 300 A current point.

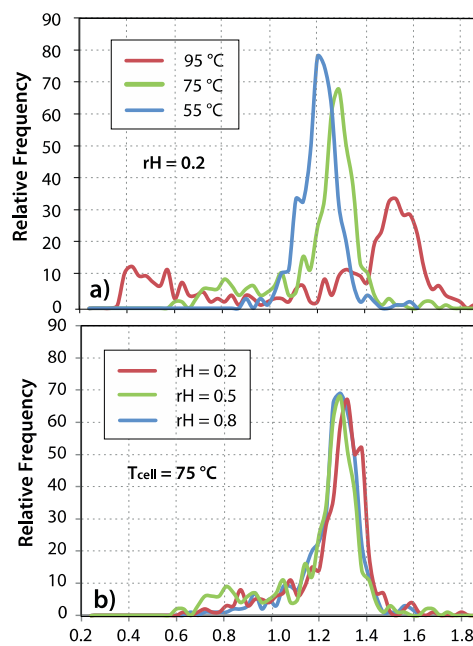


Figure 4: Current density distribution at 1.25 A/cm² (average) with $p_{gas} = 0.25$ MPa and gas stoichiometries of 1.3/1.5 (A/C); a) for the temperatures indicated and rH = 0.2; b) at 75 °C cell temperature and different gas humidities; data obtained on single cell.

For all operating conditions shown in Figure 3, the histograms of the current density distributions are given in Figure 4. The top graph shows the influence of temperature between 55 and

95 °C at a constant low rH of 0.2. Clearly the current density distribution is most narrow at 55 °C. The higher cell temperature show slightly bimodal distributions, caused by the lower current density at the gas inlets. The bottom graph shows, in line with Figure 3, that at a cell temperature of 75 °C the current density distribution is almost independent of the feed gas humidity, only at rH=0.2 a shallow tail down to 0.6 A/cm² is observed.

In summary, the current density distribution results show, that the circular flow field design with quasi-constant gas speed for anode and cathode¹⁵ generates in pure oxygen operation reasonably homogeneous current densities even for severe conditions such as 75 °C cell temperature and inlet humidities of rH=0.2. This indicates that the flow field produces an optimal water distribution and allows adequate removal of liquid water, which leads to minimal overvoltages with the given components. This may also be an advantage with respect to durability.

Durability

The high oxygen partial pressure is of concern regarding the durability. Oxygen may be directly involved in the corrosion of the catalyst support and the chemical stability of the membrane. Permeation of oxygen across the membrane is one of the causes for the formation of radicals through the hydrogen peroxide route²³, therefore higher partial pressures may lead to higher hydrogen peroxide concentrations and thus to higher radical production rates.

Durability was assessed in long-term experiments with short stacks (6 – 9 cells) in flow-through mode for the gases using the base case conditions given in the experimental section (Table 1). Two different current profiles were applied for durability

experiments, the cell voltages were sampled and averaged over the constant current period and the development of this voltage value is analyzed as function of time. The voltage decays with time, the developments are plotted in Figure 5 for different current densities for the low and high current profile. Three facts can be seen from this data set: i) absolute degradation rates in the order of 10 μV/h at a current density of ca. 0.2 A/cm² are observed; ii) degradation rates are a function of current density (losses are increasing with current) and iii) doubling the average current density does not affect the degradation rate, when comparing at the same load.

With the “high current” protocol, the cell voltage at 0.34 A/cm² is about 20 mV higher at the begin of experiment (Figure 5c) than in the low current case, which is due to better membrane humidification at the higher average current density.

The homogeneous current density distribution in the H₂/O₂ cell makes it feasible to employ the simple model of equation (1) for interpretation of the durability data. In order to assess the main source of degradation, therefore eq. (1) was used for fitting the data (the current cycling protocol makes 3 current density/voltage points available for each time step) to discriminate between ohmic and catalytic degradation losses. Table 2 shows the results for few discrete time stamps. The catalytic overvoltage rises fast in the first 1000 hours and then levels off. This may be attributed to changes of the catalyst properties (particle growth, dissolution) which is known to be fastest in the beginning of life. The ohmic voltage losses show an inverse behaviour with accelerating losses over time. This might be due to increasing mass transport losses, which are neglected in the model, and lead to an apparent increase of the ohmic losses.

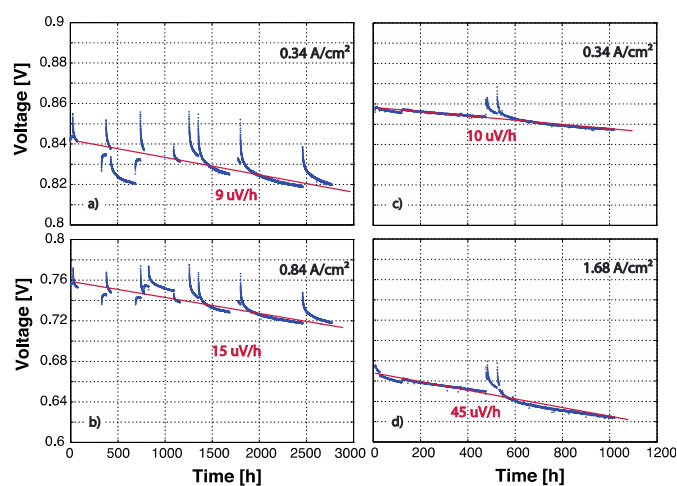


Figure 5: Average cell voltage of 3-cell sub-stacks run with current cycling protocols at 74 °C at current densities indicated; a & b) for cycling protocol “low current” 0.17 – 0.84 A/cm² (40 – 200 A); c & d) for cycling protocol “high current” 0.34 – 1.68 A/cm² (80 – 400 A).

testing (“low” up to 0.84 A/cm² and “high” up to 1.68 A/cm²) which are shown in Figure 1. During the durability

Table 2: Development of ohmic and electrocatalytic losses for 2700h test (see Figure 5a/b), as evaluated by equation (1) for the parameters *R* (Resistance), *b* (Tafel slope) and *E*_{ocv} (Open circuit). Overvoltages (*OV_R* and *OV_C*) are given for the 0.84 A/cm² point to compare the development of the losses with time.

Time stamp	Ohmic losses		Catalytic losses		Open circuit
[h]	<i>R</i> [mΩcm ²]	<i>OV_R</i> [mV]	<i>b</i> [mV]	<i>OV_C</i> [mV]	<i>E</i> _{ocv} [mV]
0	111	93	77	177	1026
100	109	91	85	196	1042
1500	114	96	95	219	1048
2300	123	104	97	223	1047
2700	132	111	97	223	1047

At conventional current densities (i.e. $\approx 1 \text{ A/cm}^2$) the degradation rates with a cyclic protocol are in a similar range as for H_2/air operation under accelerated stress conditions.²⁴ So no detrimental effect, i.e. proportionality of degradation with oxygen partial pressure, was observed. In pure oxygen based operation even some degradation might be compensated by moderate increase of the operating pressure, a compensation strategy which is much easier to realize than in H_2/air system operation.

When assuming that start/stop events do not cause higher degradation for H_2/O_2 operation than for the H_2/air case when conducted with an appropriate protocol²⁵ operational durability in the order of at least 5000 hours is expected for dynamic operation and may be longer under constant conditions.

Efficiency

Fuel-to-power efficiency of a fuel cell depends on the current density and is difficult to be judged on the cell level. Therefore, here results from a complete system are reported. For the above described technology, a stack with 178 cells has been built, tested and integrated with a compact balance of plant.

The system is designed to be operated with current densities up to 1 A/cm^2 , i.e. in a similar current range as hydrogen/air. However it is expected that the use of pure oxygen will lead to higher system efficiencies. The system, using the same stack could also be operated with current densities up to 2.5 A/cm^2 , then similar voltage efficiencies as for a H_2/air system would be expected (see Figure 2).

The system with H_2/O_2 technology (see Figure 6) differs from a conventional H_2/air fuel cell system mainly in the realization of the cathode loop. Because a pure gas is also used at the cathode, the two gas circuits become similar; gas is

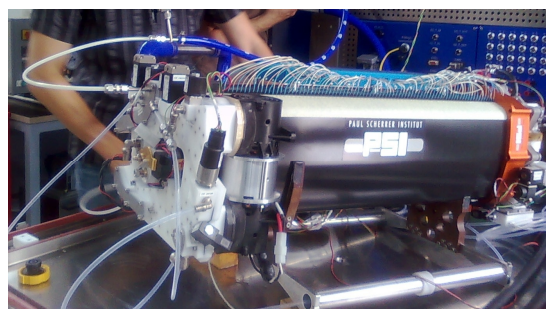
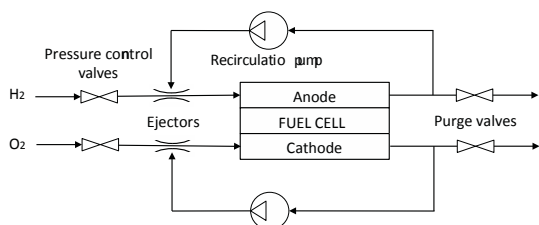


Figure 6: top) simplified system layout of H_2/O_2 system; bottom) 25 kW system with all system components (except heat exchanger) integrated in the end-plates.

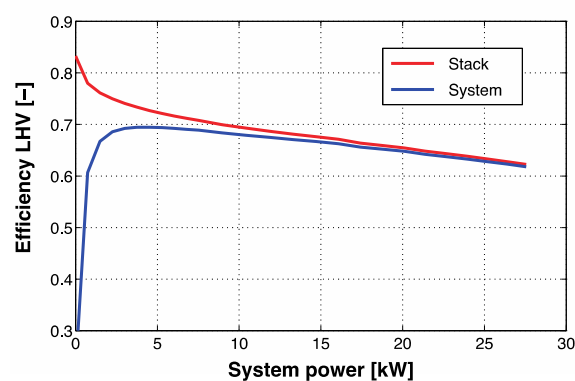


Figure 7: Stack and system efficiency of H_2/O_2 system (both 178 cells in stack) at gas pressures of 0.35 MPa.

recycled while water is separated from the loop. Because of the much smaller volume flow at the cathode than in the case of air operation, and recycling of humid gas, no additional humidification is required. Figure 3 shows that the current distribution at $75 \text{ }^\circ\text{C}$ is not sensitive to humidification. Coolant loop and other controls are comparable to a H_2/air system. The compact nature of the gas circuits allows for integrating all components (except for the heat exchanger) close to the stack, as shown in Figure 6, which illustrates a 25 kW (rated power) system based on a 178-cell stack.

For system efficiency, gas utilization is a key factor. In the pure oxygen system no nitrogen is diffusing to the anode from the cathode, so hydrogen and oxygen are only diluted by their own inert contaminants. This can be taken care of by an optimized purge strategy. Based on the measured permeation rates, the loss of hydrogen by permeation through the membrane and catalytic combustion of permeated oxygen is estimated to be less than 5 mA/cm^2 equivalent. Hydrogen utilizations of $> 99\%$ can thus be achieved at current densities above 0.5 A/cm^2 .

Efficiency data of the system is shown in Figure 7. The red curve shows the conversion efficiency in the stack (calculated with LHV of hydrogen). The average cell voltage is above 0.77 V at rated power of 25 kW, leading to a cell efficiency of 63% (LHV). In the absence of an air compressor/blower the parasitic power of the system is minimal and at rated power, also system efficiency is close to 63% (LHV). The best system efficiency, 69% (LHV) is obtained at a part load of 4 kW.

It is of interest to compare the system efficiency to a H_2/air PEFC system, however an unbiased comparison is not trivial because not all relevant boundary conditions can be kept the same for the H_2/air and H_2/O_2 systems. Same rated power, same power at a given cell voltage or same stack size can be chosen, but the other parameters will vary. In this work it was decided to compare systems with identical active area of the stack, thus systems of similar stack cost are compared.

For a comparison on the system level a reasonable maturity and optimization state of the systems is required, therefore a

commercial H₂/air system HyPEM 16 (rated power 16 kW) from Hydrogenics is compared to the prototype H₂/O₂ system described above. The two systems have stacks with comparable active area. Further system characteristics are given in Table 3.

In Figure 8a the current/voltage characteristics of the two systems are shown. In the current density range of 0.2 - 0.7 A/cm², the average cell voltage on the oxygen system is 100 - 130 mV higher as compared to the commercial H₂/air system. According to equations (2) and (3), the theoretical value for the oxygen partial pressure effect is 35 - 70 mV only depending on the value of γ in eq. (3). So the remainder of the voltage gain can be attributed to reduced ohmic and transport losses and a more homogeneous current density distribution in case of the oxygen system. If the voltage gain of approx. 100 mV would have to be realized by improvement of the catalyst activity or catalyst surface in the air system, an increase of the catalyst specific exchange current density $i_{0,S}$ (or roughness factor f_r) in the order of 30 would be required.

The higher cell voltages in combination with a low parasitic load of the balance of plant in the oxygen system also translate into a considerably higher system efficiency. Figure 8b shows the tank-to-DC system efficiency for both systems. The highest efficiency of the H₂/air system is 57% (LHV) at about 5 kW system power. At the same power the H₂/O₂ system efficiency is 69%. At the rated power of the air system its efficiency is 52%, while the oxygen system returns 67%, a relative advantage of almost 30%. With the use of pure oxygen

Table 3: Comparison of H₂/air and H₂/O₂ fuel cell system data.

Property	Hydrogenics HyPEM 16	Belenos/PSI B25
fuel	H ₂ /air	H ₂ /O ₂
Cell active area [cm ²]	500	238
#cells in stack	80	178
Stack active area [m ²]	4.0	4.2
System weight [kg]	96	74
Power density [kW/kg]	0.17	0.3

simultaneously a higher efficiency and total power are achieved with the same stack size.

If the hydrogen/oxygen system would be designed for same efficiency as the hydrogen/air system, then an about 3 times smaller stack could be employed (current density at 0.65 V would be 2.2 vs. 0.7 A/cm²). This would lead to a substantial cost reduction of the system. Possible applications, using this mode of operation are e.g. uninterruptible power systems.

Safety

Operation with pure oxygen requires special consideration of safety with respect to mixing of hydrogen and oxygen. Gases crossing over through the membrane combust completely in the opposite catalyst layer, no accumulation is observed in the recycled gas volumes during normal operation. To prevent dangerous conditions in the system from i.e. membrane failure, the hydrogen loop is operated at slightly higher pressure and the hydrogen content in the oxygen loop is monitored by a sensor. The system can shut down when hydrogen levels increase unexpectedly.

4 Conclusions

Reconversion of hydrogen, produced by the power-to-gas process, back to electricity requires the highest possible efficiencies (> 60% HHV) in order to achieve round trip efficiencies of >50% when employing the best electrolyzer technology (SOEC promises to reach 90% HHV⁷). Storing and using the pure oxygen available from the water electrolysis, allows for the development of a highly efficient polymer electrolyte fuel cell technology for this reconversion process. Therefore we have investigated the properties of hydrogen fuelled polymer electrolyte fuel cells using pure oxygen as the cathode reactant.

When operated in the conventional current density range (up to ca. 1 A/cm²), high system efficiencies approaching 70% LHV (58% HHV) are observed on the 25 kW system level. Such values are achieved through exceptionally even current distributions on the cell level and low parasitic power

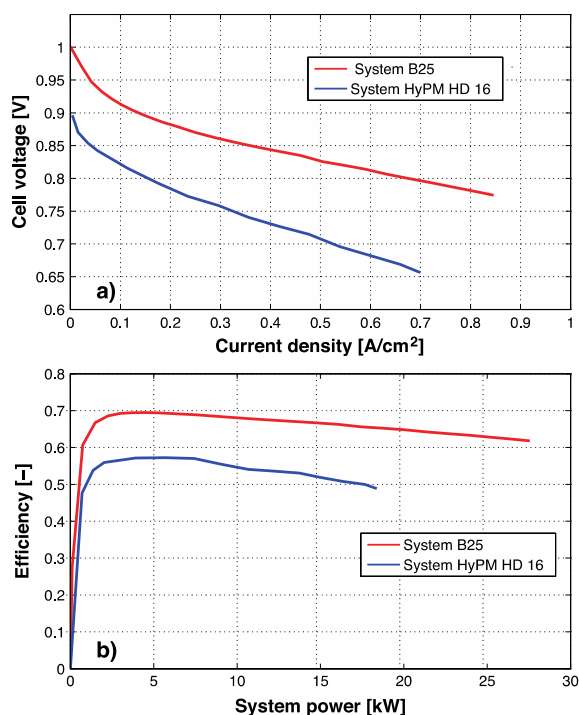


Figure 8: a) current/voltage characteristic of the H₂/air and H₂/O₂ fuel cell systems as function of stack current density; b) comparison of system efficiencies (LHV) of the H₂/O₂ and the H₂/air system (details see Table 3).

consumption in the balance of the plant. It seems feasible that the >60% (HHV) efficiency benchmark is achievable with scale-up and further development of the system components and controls, cell structures and electrochemical components.

The durability in accelerated cycling tests with pure oxygen operation was investigated in sub-stack operation and showed degradation rates of 10 - 15 $\mu\text{V}/\text{h}$ at the desired current densities, a value comparable to air operation. For the reconversion application, with operating times of > 1000 h per year, further improved durability will be required if lifetimes of at least 10 calendar years are envisaged.

The results also show that high specific current (ca. 2.5 A/cm^2) and power densities (1.7 W/cm^2) can be achieved with the H_2/O_2 technology. In this case, system efficiency will become comparable to operation with air, in the order of 50% LHV, however the required stack size is a factor of 2-4 smaller than for the H_2/air systems, offering substantial system cost reduction.

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