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Onsite hydrogen generation and hydrogen recycling for refuelling FCEVs using an electrochemical hydrogen compressor

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Summary

Fuel Cell mobility is said to be an important future technology, but it is stuck in a classical chicken-egg-dilemma. There are not enough hydrogen refuelling stations and therefore only a few cars – and the other way round. Thereby hydrogen is abounding everywhere, it just has to be made exploitable. This paper introduces two opportunities for refuelling of hydrogen independent of the existing infrastructure: generating hydrogen at home or recycling hydrogen from the exhaust gases at industrial facilities. Both opportunities are getting feasible by using a new technology, the electrochemical hydrogen compressor (EHC).

Keywords: mobility concepts, fuel cell vehicle, hydrogen, recycling, compressor

1 Introduction

For the future integration of a high share of renewable energies into the energy system it will be necessary to transfer the generation surplus into other sectors [1]. By coupling of the sectors energy, mobility and heat it is possible to reach synergy effects, for example by charging battery electric vehicles (BEV) with surplus electricity or by using the waste heat of energy processes for the heating system of a building. The usage of the power generated from renewable energy sources for the production of hydrogen using water electrolysis is currently researched mostly in a large scale [2], considering power-to-gas in combination with a wind farm. Hydrogen as an energy storage medium has the advantage that the stored capacity is just scalable by the tank volume and that the hydrogen and the energy inside can be stored for a long time. In the current power-to-gas activities the hydrogen is methanised in many cases and feed to the gas grid because it cannot be used on-site. The transport of industrial generated hydrogen to refuelling stations or direct consumers is usually realised with liquid hydrogen. This liquid hydrogen is produced by cooling, expansion and liquefaction of gaseous hydrogen. For the cooling it has to reach a temperature of about 20 K [3] which can be achieved by a two-step-process. The work for the liquefaction process is around 40 % of the containing energy [4]. Furthermore, every day the stored liquid hydrogen has a boil-off loss of 0.5 %. With the liquefaction a high energy demand and high transport costs come along. For a smaller amount of hydrogen the costs and the specific energy demand is particularly higher. The transport of the green hydrogen from the power-to-gas projects is therefore too expensive. In these small scale applications (compared to industrial produced hydrogen) the methanisation is cheaper and easier to handle. But this also means that high-quality hydrogen is polluted and loses its worth.

On the other hand hydrogen is missing for the refuelling infrastructure of fuel cell electric vehicles (FCEV). FCEVs have advantages compared to BEV because of their short refuelling times and the expanded range. Although some automobile manufacturers have already placed models at the market the demand is still low due to the missing infrastructure. The prizes for FCEV are higher than for combustion vehicles because fuel cells use platinum as a catalyst and all components are expensive due to the low volumes. For the extensive application of FCEV a well-developed refuelling infrastructure is needed. Worldwide 274 hydrogen refilling stations (HRS) are existing (January 2017, [5]), in Germany you can find 30 operating stations by June 2017. For buying a FCEV especially in the private sector you need a reliable local refuelling possibility and in the regular case you depend on only one refuelling possibility in the neighbourhood. 30 stations are not enough to reach every place in Germany with a FCEV and go back. A HRS has construction costs of around 1 to 2 million euros. At the moment there is a classical chicken-egg-dilemma. No-one builds a refuelling station when there are no cars and no-one buys a car when there are no refuelling stations.

Thereby hydrogen plays an important role in the chemical and in the metal processing industry. The water electrolysis shows the potential to generate hydrogen in a decentral way in different scales. This potential hydrogen is unused so far. If it could be made exploitable for the refuelling of FCEV then less HRS would be needed, the chicken-egg-dilemma could be alleviated and the market for FCEVs could be activated.

The electrochemical hydrogen compressor (EHC) could be the missing piece which inhibited this use until now. With an EHC a hydrogen storage and refuelling system could be also conceivable for a smaller scale. Power-to-gas in a small scale can be used directly and decentral for a surplus of locally generated electricity from photovoltaics or from other renewable energy sources. A long-time storage possibility would be provided. If required, the produced hydrogen can be used for the drive of a FCEV. The usage for the mobility ensures a consistent demand of hydrogen during the whole year which can be planned easily. The recycling of hydrogen from industrial processes would also be feasible because these processes also run quite constant during the year. With an EHC the efficiency of the hydrogen providing processes can be improved and the costs can be reduced.

2 The electrochemical hydrogen compressor

PEM-electrolyser, PEM-fuel cell and hydrogen tanks are commercially available and working fairly reliable. Whereas the compression often goes along with some challenges. Mechanical compressors are industrial standard, for example diaphragm or piston compressors. These are using the displacement principle and therefore have moving parts. An operating noise and high maintenance effort arise from that. Mechanical compressors are working rather adiabatic (Figure 1) and the temperature of the gas increases during the compression.

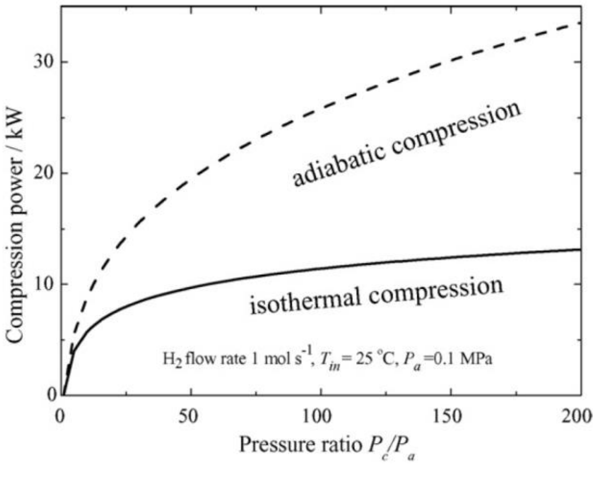


Figure 1: Comparison of adiabatic and isothermal compression [6]

The increase of the temperature of the working medium reduces the compression power of the mechanical compressor. For higher pressures often a multistage compression with an intermediate cooling has to be used. High space requirements go along with it. Further challenges result in the usage of oil in many compressors and a contamination of the hydrogen with oil particles and abrasion. The diaphragm compressor bears the risk of a tear in the membrane, which can also be a cause for a hydrogen contamination. Mechanical compressors are a good choice for high volume flows and in industrial applications where noise does not matter.

An alternative is the EHC. The EHC consists, like every electrochemical energy converter, of an anode, a cathode and an electrolyte. The electrolyte of the EHC is a polymer electrolyte membrane (PEM). Only hydrogen protons are pumped through this membrane, which is apparent in Figure 2.

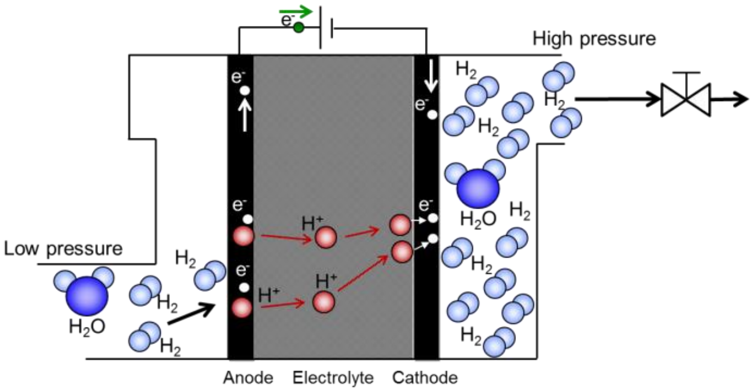


Figure 2: Structure of an EHC

When the anode and the cathode are connected by an external power source, the diatomic hydrogen is forced to split up into 2 protons and 2 electrons. The difference in potential leads to a transport of the protons to the cathode side through the membrane, where they recombine again to diatomic hydrogen. When the valve on the cathode side is closed, the pressure increases.

The electrochemical compression has the following advantages compared to the mechanical compression:

- First measurements show the opportunity of an isothermal compression [7] and therefore a better efficiency (Figure 1).
- It has no moving parts and therefore it is very robust, low-maintenance and silent.
- If the cathode outlet is closed the pressure increases. The higher cathode pressure leads to the opportunity of using wet hydrogen on the anode side, because the humidity will be driven out and the hydrogen will be dried.
- The EHC is easy scalable and can be adapted to a higher demand. As mainly only the stack will be enlarged it has less space requirements compared to the mechanical alternative.
- Only hydrogen protons are transported through the membrane, other substances remain on the anode side and a gas purification can be achieved. Hydrogen with a purity of 97.9797 % is possible [7] by a recovery rate of nearly 100 % [8].

The quality of the hydrogen is important for the durability of fuel cells. If the hydrogen is polluted from the mechanical compression the catalyst can be destroyed because it is very sensitive to external gases.

When comparing the operating temperatures 80 °C, which is also the operating temperature of PEM fuel cells, seem to reach the best efficiency (Figure 3).

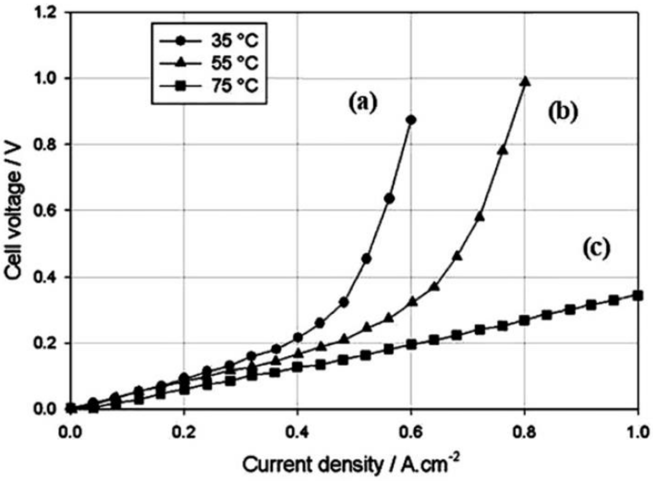


Figure 3: I-V-characteristics measured on the compression cell for different temperatures [9]

For a first evaluation of an EHC we measured single cells to validate the data from the literature. Based on this we tested different gas compositions and operating conditions. Our measurements of a single cell show that the EHC is better suitable for a high mole fraction of hydrogen. When applied to a gas composition with a lower mole fraction of hydrogen a higher voltage and resulting from that more power is needed for the compression (Figure 4).

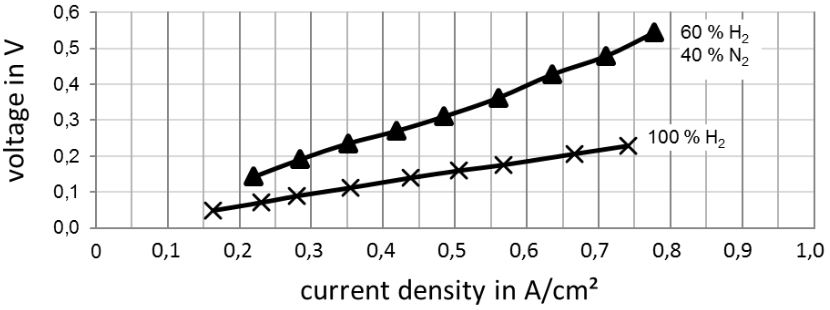


Figure 4: I-V-characteristics of a measured cell for 80 °C and rel. humidity of 80 %

The current challenges of the EHC are the dependency of the input gas composition, the tightness of the cells and the whole reliability. Some companies are already working on the development of the EHC for the market, but there is still a high research potential in the operating conditions, the costs, the catalyst coating, the best material and the optimal flow field, which leads to a prevention of the saleability of the product. Further measurements and validations are needed to better identify the weaknesses of the technology and to find the optimized operating conditions.

3 Home refuelling

In Germany 88 % of the passenger cars are in private ownership. Nearly 70 % of these could have access to a private refuelling station because they usually park in a private garage, a carport or on a private parking space. These private cars had the potential for home loading if they were BEVs. Nearly the same potential could be conjectured for the FCEV which means around 28 million cars could be refuelled at a private refuelling station in Germany.

Depending on the technology and the concept only supplies for water, grid electricity or electricity generated by renewable energy sources are needed and a little bit more space for the refuelling station compared to a wall box. First attempts come from the company Honda, which is developing its Smart

Hydrogen Station (SHS) since 2002. The current version was realised in the year 2014 in four different places in Japan to generate data and to carry out a long-time test. Honda's SHS uses a high-pressure electrolyser for the generation of the hydrogen. In a high pressure electrolyser a PEM cell is fed with high-pressure water, the product hydrogen then has already a high pressure and the additional compression can be omitted. It is estimated that the water compression causes a lower energy demand than the separate mechanical hydrogen compression (Figure 5). Another possibility is the usage of an electromotive force generated by the difference of partial pressure in the hydrogen. In the current research stadium different process variations and their potential were examined and compared [10, 11, 12].

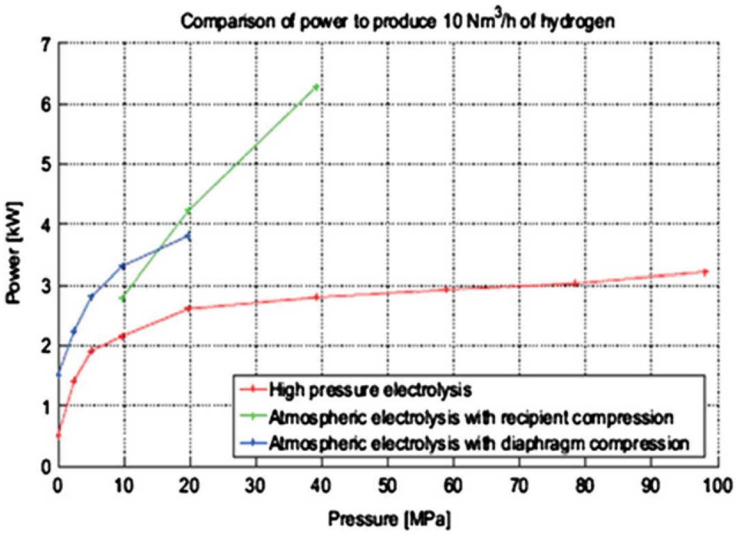


Figure 5: Comparison of the power to produce hydrogen [11]

In the literature the electrochemical compression and the correlating low energy demand for the compression are considered to be a comparable alternative to the high pressure electrolysis [7]. Therefore, we developed a concept for a hydrogen home refuelling system using an atmospheric electrolyser combined with an EHC instead of, like Honda, a high pressure electrolyser.

The EHC fits perfectly in an application in the private sector. As it makes no noise and it produces no exhaust gases it has a big advantage compared to the mechanical compression. Hydrogen can be generated by water electrolysis using a PEM-electrolyser. When electricity, which is generated by the roof photovoltaics power plant, is used for the electrolysis, the produced hydrogen is really green. The produced hydrogen can be stored in a tank for a long time; the capacity is just proportional to the tank volume. The PEM-EHC can easily be integrated between the PEM-electrolyser and the PEM-fuel cell. In Figure 6 the structure of the home refuelling system is shown.

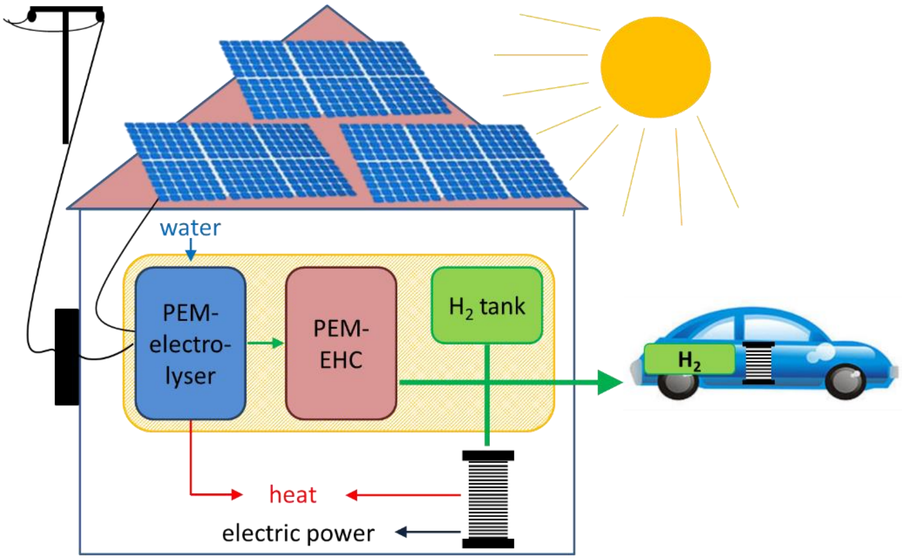
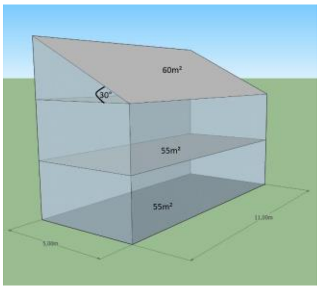


Figure 6: Schema of a home refuelling system

For the refuelling of private cars at home the refuelling time is of lower interest. That leads to the opportunity that the FCEV can be refuelled at night with a lower volume flow analogous to the loading of private BEVs.

For the simulation of the feasibility a model household (Figure 7) with the following parameters was set up:



- 3 persons
- 110 m² living space
- pent roof with 60 m² for photovoltaics (PV) usage
- installed PV power 6.76 kW
- electricity consumption per year 3,750 kWh (1,250 kWh each person).

Figure 7: Model household

For the electricity generation measured data of a photovoltaics power plant in Mannheim, Germany, were used. The load profile was taken from the VDI4655.

Combining the load and the generation leads to the residual load (Figure 8, left). This residual load very often takes on negative values during the year, which means that the electricity generation exceeds the load. This surplus of generation would be feed to the grid but could also be stored and used for the refuelling of a FCEV. The run of the storable generation surplus is shown in Figure 8 (right).

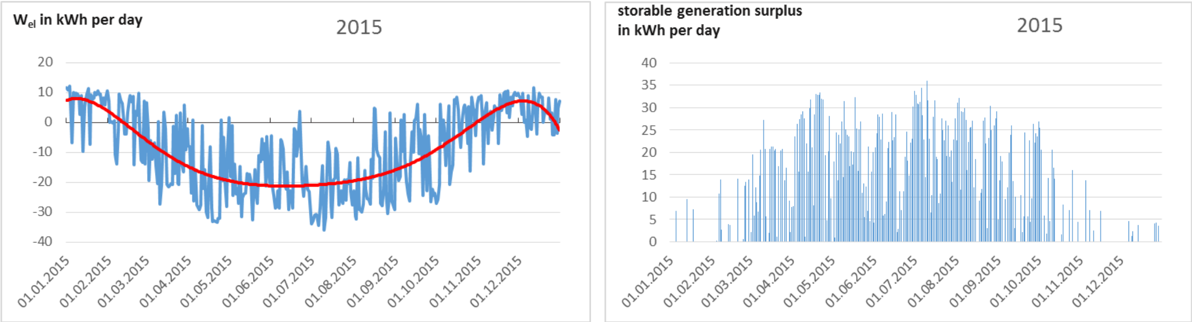


Figure 8: Run of residual load during year 2015 (left), storable generation surplus during year 2015 (right)

In the consideration of the current state the storable electricity surplus is 4,151 kWh per year. Assuming an efficiency for the electrolysis of 70 % and for the compression of 85 % around 75 kg of H_2 per year could be used for the refuelling. With 75 kg H_2 a FCEV could go for a distance of roughly 7,500 km. As the average mileage of a passenger car per year is about 14,000 km the missing hydrogen could then be refuelled at a HRS. For the future evaluation more efficient PV modules and increases in the efficiency of the electrolyser and the compression could lead to a higher refuelling amount and therefore to a higher travel distance.

Cost reductions for the system will come from less and cheaper catalysts, cheaper materials for membrane and bipolar plates. Significant cost reductions for PEM cells and therefore for PEM-electrolyser and PEM-EHC will arise by economies of scale.

The usage of hydrogen as an energy medium is an easy and extraordinary clean, environmentally friendly solution. Because only water and electricity which is generated by renewable energy sources are used the CO_2 emissions can strongly be reduced. The independency of fluctuating prices like the oil price or the gas price is also an important advantage. Additional advantage is that the temperature of the waste heat source, which is like in all PEM-technologies around 80 °C, is in a range that meets the demand of a household and could therefore be used for the heating of the building or for hot water.

Home refuelling has the opportunity to solve the chicken-and-egg problem of the FCEV branch and to push the spread of this technology forward. Then massive cost reductions will come along with it because of higher volumes. That means that the currently high costs of the whole fuelling infrastructure, the costs of the cars and the costs of all components will decrease.

4 Refuelling from hydrogen recycling

50 Mt [13] is the global annual production of hydrogen and most of it is produced and used within refineries and process industry for hydrocarbon hydrogenation. Steam methane reforming and pressure-swing-adsorption (PSA) are the methods of choice for respectively the generation and purification of hydrogen. A minor fraction is produced via electrolysis.

In most cases the hydrogen is polluted by the process. In Table 1 some gas composition for waste gas from different sources are shown.

Table 1: Gas compositions of waste gas from different sources

Source/ Molar % disregarding water	**H_2**	**N_2**	**O_2**	**CH_4**	**CO_2**	**CO**
Microelectronics industry	100 - 50	50 - 0				
Steel factory recycling	≈ 93	1	0.5	5	0.5	0.05
Ethylene plant recycling	60 - 80			20 - 40		
Biomass gasification	30 - 60			20 - 25	3 - 40	1
Biomass (SNG with residue H_2/Power2Gas)	2.2	4.3	2.1	39	51.7	0.014
Forming gas (float glass)	75	25				

After many industrial processes also traces of for example HCl, PH_3, AsH_3 or NH_3 can be found. Therefore in the first step the waste gases are cleaned of poisons by using dry bed absorption, wet scrubbing or thermal treatment. The resulting gas is usually left to the ambient air with the containing hydrogen.

Recycling of the hydrogen in the waste gas is not cost-effective with conventional methods. PSA is the current industrial standard for large scale hydrogen purification for a standard flow rate of 100 to 20,000 m^3/h. For instance PSA is used at refineries to purify reformate and at ammonia production plants to recycle hydrogen from purge gas fractions. At very large industrial scale these systems are very efficient in terms of OPEX and CAPEX for hydrogen. Since PSA is a batch process often multiple column systems are used to reach an almost steady output flow of hydrogen and to meet the purity requirements. PSA can reach a hydrogen purity of 99.999 %, but can recover only 70 – 85 % of the hydrogen. For lower flow rates PSA is not economically viable [14].

Other technologies are not able to reach a high quality of hydrogen. Cryogenic separation, polymer membrane diffusion or metal hydride separation are a good choice for a prepurification from poisons, but the resulting hydrogen has only a purity of 90 – 99 %. As fuel cells require a purity of 99.999 % according to ISO 14687-2 this hydrogen has to be cleaned further.

The palladium membrane diffusion and the catalytic purification make a purity of 99.999 % possible by a recovery rate of up to 99 %. Both techniques are very sensitive towards sulphur and also show stability problems with CO. Another disadvantage is that they have a gas selectivity for H_2 over CO_2 of 1,000 – 5,000.

Electrochemical compression has the potential to fill this gap. The PEM-EHC transports only hydrogen protons through the membrane, which also means that all other gases stay on the anode side. The hydrogen on the cathode side is purified. The EHC is more suitable for small scale purification devices but it is also scalable to bigger demands. The one-step cleaning process includes the compression of hydrogen and therefore the technology has an advantage in efficiency compared to PSA. Furthermore because of its lower required space it can be easier integrated into existing processes. A concept for a flow diagram with an integrated EHC system for the hydrogen recycling is shown in Figure 9.

The recycled hydrogen can directly and onsite be used to refuel the FCEVs of the company's carpool. It is also conceivable to refuel trucks, transport vehicles or fuel cell forklifts.

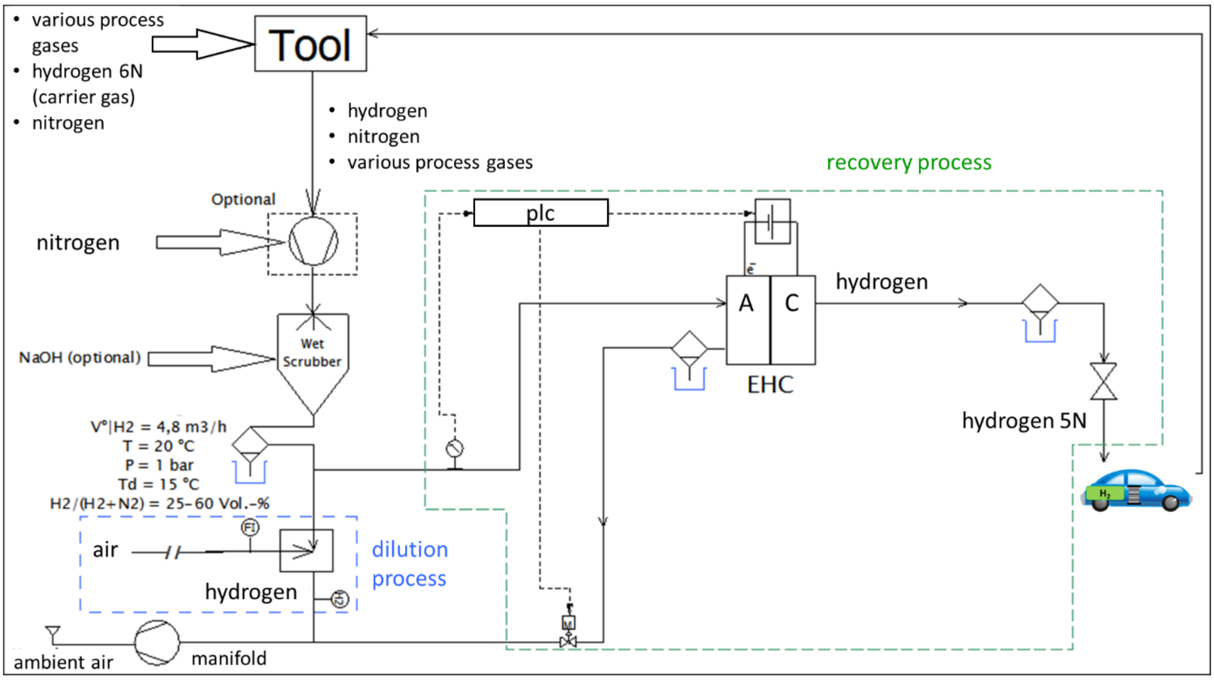


Figure 9: Concept for a flow diagram using the EHC technology for the H_2 recycling

5 Project MEMPHYS

The project MEMPHYS (MEMbrane based Purification of HYdrogen System) targets the development of an electrochemical purification system based on a membrane. For a broad application range it should be a stand-alone system based on a scalable module. Applications are for instance hydrogen recovery from biomass fermentation, industrial pipelines, storage in underground caverns, and industrial waste gas streams.

In detail, the purification process will be a two-step process. A catalyst-coated proton exchange membrane will be assisted by one selectively permeable polymer membrane. Furthermore, because the standard catalyst could be sensitive to impurities in the gas, the development of an alternative anode catalyst for the EHP (electrochemical hydrogen purification) cell, an anti-poisoning strategy and an on board diagnostic system will be developed. These measures render the MEMPHYS system multi-deployable for purification of a large variety of hydrogen sources.

Another feature of the MEMPHYS project is the ability to achieve simultaneous compression of the purified hydrogen up to 200 bar, therefore facilitating the transportation and storage of the purified hydrogen. The targeted values are summarized in Table 2.

Table 2: MEMPHYS system in numbers

Parameter	**Value**
Feed gas pressure in bar	0.05 - 100
Feed gas temperature in °C	20 - 80
Output pressure in bar	200
H_2 production rate in kg/day	5
Energy consumption in kWh/kgH_2	< 5
CAPEX in €/kgH_2/day	< 1,500

Ozone cleaning combined with on-board stack diagnostics will further enhance the flexibility of the EHP system. Detailed CFD studies will result in the optimum flow field design for the targeted > 90 % recovery rate. “Round robin” testing of stack and system at different partner facilicties will ensure a reliable assessment of system performance and economic viability. The iterative sequence of modelling, building and testing of EHP cell, stack and system components should result in a reliable validation of a small scale hydrogen purification system at relevant conditions (TRL5).

A low CAPEX for the EHP system is feasible due to the significant reductions of system costs that result from recent design improvements and market introductions of various electrochemical conversion systems such as hydrogen fuel cells.

6 Economic Evaluation

The International Energy Agency estimates in its Technology Roadmap Hydrogen and Fuel Cells from the year 2015 [15] that in the year 2050 worldwide more than 100 million FCEV will be on the road. The main markets are the US, Japan and the European states Germany, France, UK and Italy. A fast market ramp-up is expected with strongly decreasing technology costs. Further reductions will be reached by economies of scale and the synergy effects because of using the same materials and components. A strong decrease of costs especially when using the PEM technology in different stages along the process chain will come along with that.

Every refuelling opportunity leads to a local market activation for FCEV. The current problem of the non-availability of the actual existing HRS can be avoided. The HyTrustPlus project comes to the conclusion that in Germany until the year 2030 1,000 HRS have to be built to supply the target amount of 1 million FCEV with hydrogen. Depending on the scenario costs between 2.8 and 7.8 billion euros will occur [16].

In [17] the refuelling costs of HRS and the impact of different configurations are examined. The analysis comes to the conclusion that more than 50 % of the station costs are contributed by the compressor so the reduction potential is enormously.

A distribution of the hydrogen supply to many smaller decentral refuelling possibilities will reduce the demand of big HRS strongly and the saved money can be spent for the development of better and cheaper decentral refuelling stations. The decentral refuelling will enlarge the market because potential customers are independent of the regional existing infrastructure.

For the home refuelling the current costs of the system are estimated as roughly 60,000 €. The biggest part of these relate to the electrolyser, the second to the EHC (Figure 10). By higher volumes and synergy effects in the PEM technology significant costs reductions can be reached as both, electrolyser and EHC, consist of PEM-cells.

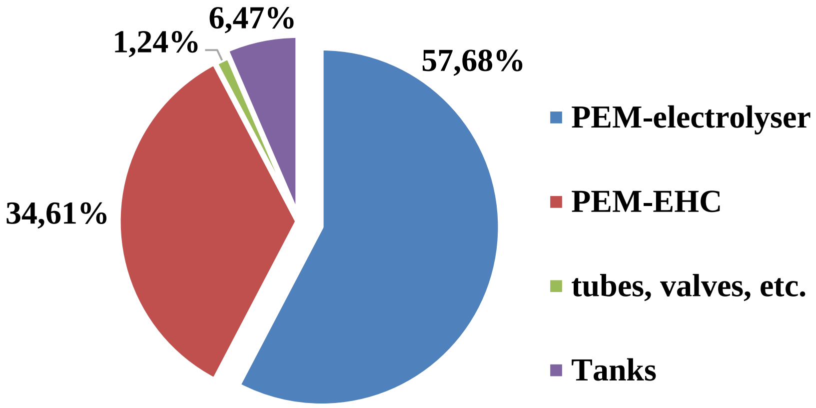


Figure 10: Current CAPEX distribution for home refuelling solution

Because of the self-generation of the hydrogen costs for buying hydrogen at a HRS can be saved. Assuming a current price for 1 kg H_2 at a station of 9.50 € savings of 712.50 € per year are possible for the calculated 75 kg H_2 per year. In the future the costs for the components of the system will decrease stronger than the hydrogen price at the station which will lead to an economically viable solution.

The recycling of hydrogen from industrial waste gases has a huge economic potential. In Germany every day approximately 4 million m^3 H_2 accrue from industrial processes. Assuming a maximum usage of this by-product hydrogen an amount of 123,000 tons of H_2 could be used for the mobility every year. With a minimum price for industrially produced hydrogen of 1 € per kg H_2 an economic value of 123 million € per year results. If the hydrogen would be sold directly onsite at the factory sites for refuelling of vehicles a price of up to 9 € per kg H_2 would be achievable. This also seems interesting for the refuelling of fuel cell trucks in an industrial area.

The cost structure for the recycling solution is shown in Figure 11. Half of the costs are related to the EHC. The special recycling module (which also consists of a membrane with a different coating and catalyst) and the valves are also big parts.

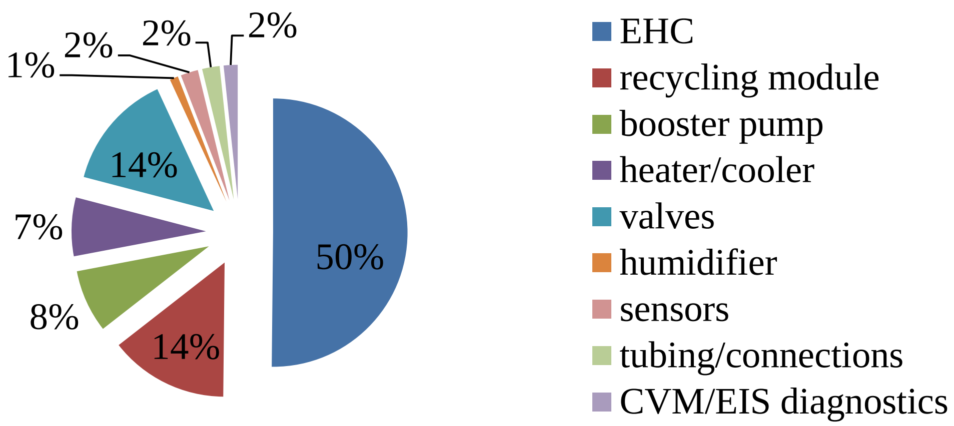


Figure 11: Current CAPEX distribution for recycling solution

When scaling up the volume flow and associating the EHC system the percentage of the costs for the EHC will increase. The surrounding system components will not increase in the same proportion than the EHC stack.

Further considerations of the development especially of the future costs are still pending. We assume that the decreasing technology costs will make the EHC also suitable for more applications like for example the recirculation in fuel cell systems or purification of hydrogen from caverns.

Acknowledgments

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Authors

Linda Schorer has a master's degree in Business Engineering. She is familiar with most energy related topics and started her PhD in October 2015 about a techno-economical evaluation of home refuelling concepts based on hydrogen using an electrochemical hydrogen compressor. For the technical investigation she is building up a high pressure storage system in the laboratory. Since January 2017 she is working in the MEMPHYS project where she is responsible for all economic considerations, different project management tasks and the organization of events, public relations and dissemination.

Professor Dr.-Ing. **Sven Schmitz**, started his career in the Volkswagen Group Research at the Technology Centre for Electric propulsion. Here he was involved resp. supervised the development of fuel cell propulsion systems for several generations of VW fuel cell vehicles. Since 2013 he is professor at the DHBW for Mechatronics and Electro Mobility and the head of the laboratory for electrochemistry. This laboratory has been built up since 2013 and covers test rigs for electrochemical cells and stacks (fuel cells, electrochemical compressors and electrolysers) ranging from 100 W up to 10 kW. Sven Schmitz is the project coordinator of the MEMPHYS project, which is founded by the FCH 2 JU.