3rd Conference on Production Systems and Logistics

The Contribution Of New Production Technologies And Circular Economy Towards Meeting The Future Demand Of Proton-exchange membrane Fuel Cells – A Literature Review

Patrick Alexander Schmidt[1]
[1]*Fraunhofer Institute for Machine Tools and Forming Technology IWU, Reichenhainer Straße 88, 09126 Chemnitz, Germany*

Abstract

The energy and mobility sectors contribute significantly towards the global CO_2 emissions. The proton-exchange membrane fuel cell finds application in both sectors and represents a possible green and sustainable technology for electricity generation. Current production rates do not satisfy the predicted demand for proton-exchange membrane fuel cells as the diffusion of this technology keeps increasing. Nor does the per-part cost guarantee a globally sufficiently broad application. The industry must overcome technological and economic obstacles to enable higher production rates at a lower cost per unit.

This research gives an overview of current proton-exchange membrane fuel cell production and stacking technologies and provides an outlook on processes that need to be improved to enable faster and lower-cost production. Additionally, the impact of remanufacturing as an end of life option on the circular economy, production, and ecological impact of proton-exchange membrane fuel cells is examined.

The knowledge generated by this research shall support increasing proton-exchange membrane fuel cell production rates to catch up with the predicted demand. Since current research on proton-exchange membrane fuel cell remanufacturing is rare, findings on this topic will support the industry in preparing for circular production processes in the future. Results of the present work include an overview of the current state of production for proton-exchange membrane fuel cells, the areas that need improvement, and the role of a circular economy.

Keywords

Circular economy; End of life; High-rate production; Proton-exchange membrane fuel cell (PEMFC); Remanufacturing.

1. Introduction

With the Paris Climate Agreement, 194 countries and the European Union set themselves to limit global warming due to increasing greenhouse gas emissions to well below 2°C and ideally to below 1.5°C [1]. Germany was responsible for 1.9% of global CO2 emissions in 2019 [2]. In 2019, Germany caused 39% of its CO2 emissions by electricity generation, 23.5% by industry, 21.5% by transport, and 16% by the building sector (only includes heat production, other sources count towards electricity consumption) [3]. In order to counteract the climate crisis, the areas mentioned above need a transformation. One possible technology for reducing CO2 emissions in the transport, industry, and building sectors are proton-exchange membrane fuel cells (PEMFC) combined with green hydrogen. The use of PEMFCs can replace the use of fossil fuels in

DOI: https://doi.org/10.15488/12172

transport and, when installed in buildings as combined heat and power units, generate electricity and at the same time use waste heat efficiently.

Due to limited distribution, the current annual production volume of PEMFC is still shallow. However, there are signals from politics, such as the National Hydrogen Strategy [4], and the economy, which point to an enormously increasing sales market for PEMFC. The Hydrogen Council forecasts that 10 to 15 million cars and 500,000 trucks worldwide will run on fuel cell technology in 2030, and 400 million cars and 15 to 20 million trucks in 2050 [5]. The Japanese car manufacturer Toyota states that its fuel cell cars production will increase tenfold from 3,000 vehicles per year in 2021 [6]. Optimistic forecasts see growth to up to 1.8 million hydrogen vehicles in Germany in 2030. The use of PEMFC is not limited to passenger cars. The technology benefits public transportation with regional trains [7].

In order to meet the anticipated demand for fuel cells, a high-rate production is required (explained in section 3.2). Furthermore, a holistic consideration of the life cycle of the PEMFC is necessary to exploit the sustainability potential fully. Therefore, in addition to manufacturing, it is necessary to examine the aspects of disassembly and recycling of the PEMFC. This paper aims to overview the existing processes for assembly, high-rate production, disassembly, and end of life (EOL) options of the PEMFC and derive necessary developments. The following section deals with the mode of operation, the components, and the application areas of the PEMFC. Subsequently, existing stacking concepts for the PEMFC, possibilities of a high-rate production, and effects on the production costs will be examined. Finally, possibilities and obstacles in the dismantling and recycling process are presented.

2. Operating principle

Fuel cells convert chemical energy from gases or liquids into electrical energy and release heat during this process [8]. In the PEMFC, this process occurs through the oxidation of a fuel and the reduction of an oxidant [9]. The fuel is hydrogen, and the oxidant is atmospheric oxygen. The membrane electrode assembly (MEA) separates the hydrogen and oxygen streams. The MEA consists of a semi-permeable membrane that carries an electrolyte and two electrodes with a catalyst layer [10]. The separating membrane is only permeable to the hydrogen protons produced during oxidation on the anode side of the catalyst but not to the electrons. It is therefore referred to as a proton exchange membrane (PEM). The flow of electrons takes a diversion via an external circuit to the cathode and generates electricity in the process. The electrons are then absorbed by the atmospheric oxygen at the cathode, forming water.

In addition to the PEMFC, there are other types of fuel cells. These differ in the electrolytes used, the fuel for the anode, the level of power, the operating temperature, and the electrical efficiency, resulting in different areas of application [9].

The application areas of the PEMFC can be divided into stationary, portable, and mobile applications. Applications in the portable sector include a power supply for small consumers. Stationary power generation up to the megawatt range and combined heat and power generation are other application areas [11]. Mobile uses for the PEMFC are trucks, submarines, cars, and buses [9]. The PEMFC is well suited for mobile applications, as the power output can be controlled dynamically [12]. This study focuses on the PEMFC.

3. Assembly

3.1 Stacking concepts

The voltage of one fuel cell amounts to 1V approximately. Most applications, however, require higher voltage. Therefore, several individual cells, consisting of alternately stacked bipolar plates (BPP) and MEA, are combined into a stack. This process can be performed manually or automated and must be prepared in previous work steps further explained in the following [13].

The MEA is the most crucial fuel cell component and consists of two components, which are the catalyst-coated membrane and the gas diffusion layer. The decal process is a method to produce the catalyst-coated membrane [14]. In this process, the catalyst material (a mixture of carbon substrate and platinum) is first applied to a carrier medium (decal) and transferred to the polymer membrane in a further step by hot pressing. To produce the gas diffusion layer carbon paper or fabric is needed. It is made from cut carbon fibres and subsequently impregnated and graphitised. A microporous layer is subsequently applied to increase the water balance of the electrode. The finished gas diffusion layer is then glued to both sides of the catalyst-coated membrane by hot pressing and cut into shape as a MEA. The BPPs are still missing for a complete fuel cell. They can be made from coated metal or a graphite composite material. While the composite plate can already be provided with a seal after the manufacturing process, the metal variant requires further work steps beforehand. The half-plates must first be cut out, which is usually done by laser, and then two half-plates must be welded on top of each other to form a complete BPP. A gasket is applied to the combined plate after a leak test. The gasket serves as insulation between BPP and MEA. This concludes the preparation for the stacking.

The stacking process starts with the lower endplate and the lower current collector. A BPP with a gasket and a MEA are stacked on top of each other alternating. This process is repeated until the desired number of fuel cells has been reached. Then, the upper current collector and the upper endplate follow, in which the media supply lines are also located. The individual components must be placed as precisely as possible. In the next step, the complete stack is pressed together to connect everything tightly and minimise the resistance of the contact surfaces. It is essential to ensure that the pressing force is applied evenly everywhere during this process. If this is not the case, significant reductions in performance and service life cannot be ruled out. The compressed state must be guaranteed permanently. The stack is held together by either tension bands or tension rods. Subsequently, an initial leak test takes place, such as pressure drop and flow tests. Before the stack is placed in a housing and covered by the distributor plate, final work steps are necessary, such as attaching the contacts and current busbars and the cell voltage monitoring module for the cell voltage. The so-called running-in on the test bench follows, where all mediums (hydrogen and oxygen) are connected. Through this process, the stack's performance can be determined. Finally, another leak test may be carried out, after which the fuel cell is installed with other system components and is ready for use [14].

3.2 High-rate production

The stacking process described in the previous section can be carried out in various ways. While the stacks are still partly stacked by hand, partially or fully automated processes have been developed in the meantime. Automated processes use carousel devices or conveyor and feed systems, although manual work steps are still necessary in some cases. Pick-and-place robots could automate these processes [15]. The extended use of robots would be a first step in the direction of high-rate production. A crucial development, especially regarding the capacities, is required in the future [16].

In the context of this paper, the term "high-rate production" concerning the fuel cell means an approximation to the production figures from the automotive industry. The cycle time for a vehicle is 60 seconds on average. In order to adapt the fuel cell stack production to this, ten cells must be stacked in one second, based on a stack with 400 to 600 cells. That situation, however, assumes that 100% of cars plant’s production need a fuel cell stack and is therefore relevant for highly specialised fuel cell production plants. If a supplier wanted to produce, for instance, 20.000 fuel cell stacks a year with 400 cells per stack, the cycle time would be roughly about 2.5s per fuel cell (assumptions: 240 days and 24h of work).

Nevertheless, the cycle times are still insufficient for the envisaged demand, so parallelisation and modularisation of the production capacities are necessary. A modular approach might allow to adapt steadily to increasing demands. Irrespective of this, production volumes must be increased to meet the planned future sales volumes of fuel cell electric vehicles (FCEVs), and new and faster production facilities must be found as a result. The necessary production capacities entail several prerequisites that need consideration as early as the planning stage of the plants. The quality and the safety of the fuel cell must not be reduced by shorter cycle times. Precision and repeatability are essential, and the handling of sensitive components must also be guaranteed in terms of quality, even in automated production. Some of the components, such as the MEA and gaskets, are flexible and thin, complicating the automated handling even further [17]. Since the same components from different suppliers can differ in shape, size and tolerances, machines must be adaptable. The same applies to the produced stacks, which have different dimensions depending on performance. As a result, it is advantageous if machines already have a high degree of adaptability, which prevents long conversion times or even breakdowns. For a smooth production process, bottlenecks in the components must be taken into account and calculated in advance. Subsequently, the cycle and throughput times must be adaptable [16].

In principle, however, making only the stacking process faster does not suffice to reach the desired production capacities. The individual component production must be improved, too. The production of BPPs and MEAs and the connection of the two components has great potential. The components could be produced in the roll-to-roll process in the future. In this process, the material is unwound from a roll, processed, and then rolled up again, which speeds up further processing and makes it possible to produce complete cells in a short time and separate them. Hence, they are directly available for the production of stacks [18]. However, even if the individual components are modified, it is necessary to observe several criteria to ensure that the fuel cell meets all requirements. These requirements are mainly regarding the performance of the fuel cell stack and system, where, among other things, thermal and electrical resistance must be minimised, but the conductivities must be equally maximised. Mediums such as hydrogen and oxygen must also be supplied and discharged without leakage, particularly when ambient temperatures change.

Furthermore, low masses and dimensions are advantageous. In addition to these aspects, manufacturing properties and environmental influences are also relevant, in connection with which material selection, manufacturing processes, and recycling possibilities are to be mentioned [19]. These criteria are described in more detail in section 4. Currently, there are still some hurdles to overcome to fully automated production. New processes with a direct coating of the electrode onto the membrane or gas diffusion layer are necessary. The production time can be significantly reduced with a continuous lamination process. Faster and, above all, defect-free production methods must also be planned for BPP, regardless of the material. In general, the quality factor plays an essential role in both the components and the final products, which is why the final controls or the methods for checking them must also be improved and accelerated [20].

Academia and industry are aware of the necessity to increase stacking velocity and stacking scalability. Research is performed on robotised high-speed stacking processes that aim to increase stacking rates through gripping several layers of cells at once or through the inclusion of in-line quality control to further reduce cycle times [21,22]. Researcher designed an automated workstation layout for fuel cell stacking through the parallelisation and decoupling of process steps [23]. Stacking velocities of approx. 2s per stack was reached. An outlook was given that cycle times below 2s are possible but would require high development costs. An increase in production capacities also has a positive effect on the production costs, as explained in more detail in the next section.

3.3 Cost factors

Even though the costs for PEMFC systems have fallen by about 50% in the last 15 years, they are still too high to achieve a breakthrough for the PEMFC as an alternative on the vehicle market alongside batterypowered electric vehicles. The high costs are expected to decrease in the future due to increased demand and the associated higher production volumes [24].

In principle, the costs for the complete fuel cell system consist of the fuel cell stack costs and the costs of the system components (for instance, air compressor, water separator, and hydrogen filter). The focus in this work is on the stack and its components since in this area, more components are highly dependent on the absolute production number. For small production numbers (1,000 stacks/year), five components are responsible for over 80% of the costs. If the projected production figures of 500,000 stacks/year are reached, only two components (BPP and catalyst) are responsible for 66% of the costs. Therefore, the total price can be reduced most effectively by saving costs through an increased production volume [16]. According to a report by the U.S. Department of Energy (DOE) from 2018, the price of a fuel cell system per kilowatt net power (kWnet) should be reduced to \$30 in the future (based on an 80 kWnet PEMFC and production quantities of 500,000 stacks/year). As an overview, the DOEs findings on the costs of the individual components, depending on the annual production volume, are shown in Table 1.

Table 1: Influence of production volume on the costs of components and the entire stack [24].

Yearly production rate of	1,000 stacks	10,000 stacks	100,000 stacks	500,000 stacks
BPPs	1,554\$	486\$	404\$	388\$
MEAs	6,546\$	2,320\$	1,121\$	915\$
Other components	1,278\$	700\$	135\$	127\$
Cost complete stack	9,533 \$	3,504\$	1,722\$	1,479\$
Cost complete stack (per kWnet)	119.16\$	43.8\$	21.52\$	18.49\$

However, these prices cannot be achieved by increasing capacity alone, which is why the production methods and the materials used must be changed [18]. For BPP, various materials and the production variants required were investigated in terms of the production numbers and costs required. Hydroforming (internal high-pressure forming) is more cost-effective than progressive stamping for large production volumes when using stainless steel. If several panels are produced simultaneously, fewer assembly lines are required, which directly impacts costs and processing time. Another way to decrease cost by changing the material is using an alternative stainless-steel alloy. A cost reduction of \$0.13 to \$0.21 per kW net could be reached, while there are no disadvantages in use.

Further savings are also possible by using a different plating process. Even if cost reductions are possible by reducing the platinum content and increasing the power density, prices still depend on the material costs [25]. Especially regarding precious metals such as platinum, the material price must be paid regardless of higher production numbers. For this reason, fluctuations must always be taken into account, and the use of other materials should also be considered if necessary.

4. Disassembly

After a defect, caused, for example, by impurities in the MEA, disassembly of the PEMFC is a prerequisite for reuse or further use. In principle, the process occurs in reverse order to assembly so that peripheral components on the stack are removed first (e.g., cell voltage monitoring unit and cooling fan). In the next step, the bracing is loosened. Then the endplates and the current collectors can be dismantled to reach the individual cells. The cells are lifted off one after the other and can then be disassembled into their components (BPP, gasket, gas diffusion layer and MEA) [26]. However, separating the sensitive parts bears the risk of damaging the components and making them unusable.

Similarly, it is hardly possible to replace individual cells, and inaccurate contact surfaces can lead to significant performance losses. For this reason, the introduction of a conductive intermediate layer between the BPPs was considered, which would also make it easier to detach them from each other. Flexible graphite could be used for this purpose. It also compensates for the poorer surface quality of the plates due to its deformability, thus reducing production costs and ensuring better current transmission. At the same time, different loads can be better compensated, which has a positive effect on the lifetime of the entire stack [27]. A similar patent from the Toyota company involves the use of an adhesive intermediate layer of fluoroplastic or silicone resin that can be detached by heat and serves as a seal. This method can also be used for other types of fuel cells and is also intended to facilitate the dismantling process. For this purpose, heat is applied to the adhesive layer and pressed apart with the help of a wedge, which makes it easier to apply the heat. In order to be able to remove residues of the adhesive better and accelerate the separation process, a heat-dissipating agent is applied along with the adhesive layer, which causes the dissolved adhesive to contract again [28].

With the help of these ideas, a reduction of the disassembly time is possible, which proves to be especially important with increasing numbers of FCEVs in the future. It becomes clear that these aspects must be considered before assembly during the planning phase of the stack. Hence, new design guidelines must be considered and used. "Design for disassembly" (DfD) is an approach that aims to simplify as well as speed up the disassembly process, make it more cost-effective, and recycle as many materials and components as possible. The right choice of materials, connecting elements, and the product's design are fundamental steps in the development. The disassembly process can only be improved with sufficient knowledge about the developed object's structure, its use, and the physical and technical limits of the disassembly process [29]. Another approach is the so-called Design for Remanufacturing (DfRem), where several design guidelines for products are followed to facilitate the EOL phase [30–32]. These guidelines include enabling easy, non-destructive disassembly through accessibility, modularity, ease of cleaning and handling, designing for multiple life cycles, resistance to wear and tear, and considering the EOL phase already during the product development process [29,30,33,34]. The DfRem method is primarily used to improve the possibilities for closed-loop capability, which will be defined in more detail in the next chapter.

5. End of life options

5.1 Proton-exchange membrane fuel cell remanufacturing

For the PEMFC to be justifiably described as a sustainable technology, it is necessary also to consider the end of the product life cycle. It can only be used in a genuinely sustainable manner if a circular economy exists in addition to the use of green hydrogen. After focusing on dismantling as a prerequisite for recycling the PEMFC in the last chapter, the following section looks at various EOL options, i.e., possibilities for returning the PEMFC to the product cycle at the end of its life cycle.

The various EOL options differ in several aspects. A distinction is made whether a product is reused in the same application or with a different purpose [33,35]. Furthermore, there is a difference in the point at which the product is reintroduced into the life cycle [36]. The re-entrance point influences the amount of lost energy and materials [37]. EOL options in ascending order of lost materials and energy are Reuse/repair, remanufacturing (product recycling), recycling (material recycling), energy recovery and landfill disposal. Given the cradle-to-cradle approach no waste should be produced. Thus, the first options mentioned should be preferred, and the latter avoided. Next, remanufacturing as a possible EOL option for the PEMFC will be discussed in more detail.

Remanufacturing, or product recycling, is the reprocessing of a product after its use phase to the quality level of a new product [30]. The exact process steps of remanufacturing are variable depending on the application but can be described as follows: Old part procurement, testing/sorting, cleaning, refurbishment, reassembly and a final test [31,36,37]. Various studies of use cases show that remanufacturing influences costs positively and, in particular, decreases environmental impacts compared to new production [36,38]. Remanufacturing results in less dependence on critical raw materials, advantages for the user due to lower prices and strategic advantages for the manufacturer [36]. Examples of remanufacturing applications in the automotive sector include engines, gearboxes, starters and turbochargers [36]. From the field of business mathematics, there are many publications on remanufacturing [39,40]. However, there is a lack of a deeper consideration of remanufacturing for PEMFC from a production engineering perspective. In this consideration, it is crucial to take a holistic approach where economic, environmental, and technological factors are considered simultaneously to develop the optimal process. Many obstacles exist that give the remanufacturing process its complexity. These include low volumes, uncertainty about the number and condition of EOL parts, increasing product complexity and disassembly as a cost driver due to manual processes and low product know-how [34,36,38,41,42].

5.2 Component recycling

While product recycling for the PEMFC has hardly been practised so far, there are already established processes for recycling at the material level, with which different materials of the PEMFC can be recovered. For this purpose, the stack must be broken down into its components. Peripheral components, such as control electronics, can be recycled via conventional e-waste [13]. Purely metallic components such as tension rods, current collector plates and end plates are further processed via metal scrap [43]. Other specific processes exist for the individual components of the fuel cell, which will be discussed in more detail below.

There is a strong focus on the recycling of MEA [13]. One reason is that fuel cell defects can often be traced back to the MEA. During the fuel cell operation, the formation of pores or the accumulation of impurities from the fuel might damage the membrane in the MEA [43]. Another reason is the material cost. About 42g of platinum is installed per stack, which is why the recovery of the materials has a high financial incentive [44]. Furthermore, the dissemination of FCEVs will lead to a sharp increase in platinum consumption, as the needed amount of platinum is ten times higher than in vehicles with internal combustion engines [44].

Processes for recovering precious metals from vehicle catalytic converters of internal combustion engines cannot be used for the PEMFC, as toxic hydrogen fluorides are produced during the combustion of the electrode [43]. Therefore, the catalyst is recovered by chemical extraction [45]. Compared to the catalyst, the recovery of the membrane has not been in focus so far but is becoming significantly more relevant because of increasing production numbers. The recovery of the membrane is complicated because it merges with the gas diffusion layer and the catalyst layer during the operation of the PEMFC, which is why the economic benefit of recovery still needs to be examined in detail [13].

Recycling of BPP depends on the material used. Metallic BPP can be recycled via ordinary metal scrap [13,43]. Graphic BPP made from thermoplastics can be cleaned and reprocessed into granulate for BPP production by injection moulding. In the case of BPP made from thermosets, re-melting is not possible, so only further use as, e.g. filling material is possible [13].

6. Conclusion

The production of PEMFCs is expected to grow, as they represent a sustainable alternative to existing technologies in several application areas. Various developments are still necessary to ensure that the diffusion of PEMFCs is successful in the future.

The current production rate of PEMFC is not sufficient to meet future demand. Steps towards high-rate manufacturing have already been taken using pick-and-place robots, carousel devices, feeding systems, and direct coating of the electrode by the roll-to-roll process. The mentioned measures, however, are currently not yet sufficient. There is still potential for optimisation in the production of the individual components and the necessary function tests. Parallelisation and decoupling of process steps are necessary to increase production rates. Furthermore, production costs must be reduced in the future, partly because the fuel cell costs make up too large a proportion of FCEVs. As production numbers increase, more cost-effective manufacturing processes can be applied, among other things. Existing EOL options and dismantling processes also need to be improved to increase the recyclability of PEMFCs. Inadequate dismantling processes hamper remanufacturing. These processes constitute a significant cost factor in remanufacturing due to mainly non-uniform and manual processes. If dismantling is considered in earlier life cycle phases, through DfD and DfRem, process costs will decrease. Increased recyclability reduces environmental pollution and dependence on critical raw materials.

Fraunhofer IWU's research efforts will address several of the problems mentioned. For example, there are already concepts concerning the component design of individual components and about high-rate technologies and manufacturing processes for producing these components. The degree of automation can be increased with robot-based manufacturing processes. However, alternative technologies are required concerning the desired cycle times of several cells per second. In this regard, there are already initial industry-oriented solutions that enable such production rates through continuous flow processes. For the further qualification of these processes, it is necessary to implement corresponding test facilities to carry out quality-relevant and cycle-time-specific optimisations using the demanding components of the fuel cells. Other innovative concepts about fuel cell design with adaptive assembly elements intend to increase the efficiency of the fuel cell in operation, especially with fluctuating ambient temperatures. These also need to be further qualified regarding an automated high-rate production.

Furthermore, the economic advantages of remanufacturing the PEMFC as an EOL option are investigated. The necessary steps to improve the EOL processes and cycle capability of the PEMFC will be analysed. Another research topic is the standardisation and automation of the PEMFC disassembly process.

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Biography

Patrick Alexander Schmidt (*1993) M. Sc., is a research fellow in the body in white, assembly and disassembly department at the Fraunhofer Institute for Machine Tools and Forming Technology IWU. He has been a research fellow at Fraunhofer IWU since 02.2020. In 2019, he completed his master's degree in industrial engineering with a specialisation in mechanical engineering at Chemnitz University. Previously, he studied industrial engineering at Pforzheim University, with an exchange semester at the Tecnológico de Monterrey, Mexico. His research areas include PEM fuel cells, disassembly processes, circular economy, and remanufacturing.