

HYDROGEN FUEL CELL VEHICLES

Swapnil Namekar¹, Harshdeep²

¹Assistant Professor, Department of Electrical Engineering,

²Student, Department of Electrical Engineering,

Bharati Vidyapeeth Deemed University College of Engineering Pune, India

Abstract- Fuel cell vehicles should be further improved. Key issues are cost reduction; higher power density of the primary energy converter, the fuel cell; wider operation ranges and improvement of operation parameters, e.g. higher operation temperature and starting ability in freezing conditions. Using advanced materials and construction principles is a key factor by meeting these requirements. The paper gives a short introduction to the technology of fuel cell vehicles and the most prominent fuel cell type for traction applications, the polymer-electrolyte-membrane fuel cell (PEFC). Progress in material development of a core component of the PEFC, the bipolar plate is described. In the second part of the paper some ideas are presented, in which way material research could help to enable suitable on-board storages for hydrogen. Namely, a new approach to design compressed gas storages and new developments in materials for solid state hydrogen storage are brought to attention.

I. INTRODUCTION

The fuel cell is in the transformation from chemical energy to electricity a very promising primary energy converter for automotive propulsion due to their high efficiency and ultra-low emissions. The polymer-electrolyte-membrane fuel cell (PEFC) - among the different types of fuel cells - is almost exclusively discussed for applications in traction because of their rugged design and suitability for dynamic operation. Therefore, this paper deals exclusively with PEFC technology. In comparative views with other vehicle power trains "tank to wheel" lowest CO₂-emissions for vehicles with fuel cell power trains were obtained with the PEFC-fuel cell technology [1]. However, extending the view to "well-to-wheel" it becomes apparent that the advantage is getting smaller or – for unfavorable fuel supply chains – CO₂-emissions could be also higher. The PEFC's preferable fuel for is hydrogen. As fuel up-to-date almost exclusively hydrogen is used, because it has been found that the realization of gas generation systems, which convert

hydrocarbons to a hydrogen rich gas on-board, is very complex [2]. Consequently, the above cited potential can only be assessed, if satisfying answers to the questions of hydrogen production, infrastructure and storage are found with regard to economics. Furthermore, technical progress is needed in fuel cell propulsion technology. Main issues are: cost of the power train; lifetime of the core components, namely the fuel cell stack; cold start ability; performance under freezing conditions; and operating range of the vehicles. Improved materials are needed to meeting the envisaged targets. This paper gives some examples for challenges in material science developing advanced PEFC-stacks and advanced hydrogen storages.

II. FUEL CELL POWER TRAIN

Fig. 1 shows the scheme of fuel cell power train applied to an electrical drive train. Torque for traction is provided by an electrical engine, which is usually fed with electrical energy by an inverter. The primary energy converter, which provides the electrical energy, is a fuel cell system. The hydrogen, which is consumed by the fuel cell, is stored in a hydrogen storage.

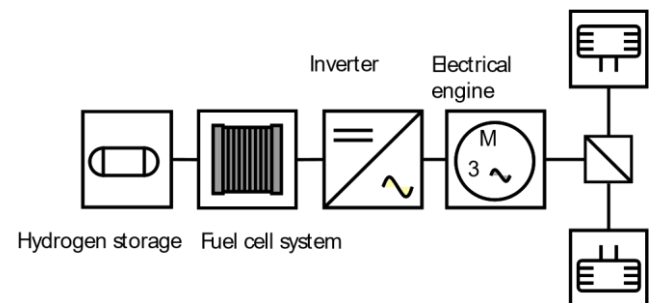


Figure 1: Scheme of a fuel cell power train.

Despite of the hydrogen supply the fuel cell system needs more sub systems: an air supply system, a heat and water management system and a control system.

However, the most important component, which determines the characteristics of the fuel cell system to a wide extent is the fuel

cell or fuel cell stack itself. Although the fuel cell technology has already made substantial improvements in the past; the road map of fuel cell developers foresees significant improvements e.g. cost reduction, durability, lifetime and power density until 2010. For instance, in the road map of Ballard the cost target for a fuel cell stack by 2010 comes to 25 USD/kW net EOL compared to 103 USD by 2004 [3].

With regard to the hydrogen storage the target of the DOE in the United States a cost reduction to 6 \$/kWh of stored energy at a recoverable hydrogen storage capacity of 4.5 wt% shall be reached by 2007 [4].

III. PEFC STACKS

A fuel cell stack has to provide all manifold functions. Fig. 2 shows on the left side a schematic representation of a single PEFC, whereby flow of media, heat and current is indicated. This scheme is not complete and some flow directions may differ depending on specific cell designs and/or specific operating conditions. The center component of the PEFC is the proton exchange membrane, which separates the two reaction layers. The combination of the membrane and the two electrodes is often called membrane electrode assembly (MEA). On the anode fuel is oxidized, whereby electrons are dragged to an external circuit and protons are conducted through the membrane to the cathodic side. On the cathode oxygen is reduced and combined with protons from the membrane and the electrons from the external electric circuit to water. Heat is released in several reaction steps and must be removed from the location of its genesis transferred to a heat transfer fluid (htf). The heat transfer fluid flows in a cooling plate, which may serve as the so called bipolar plate (BPP) as well. The function of the layers - gas distributor and gas diffusion layer - is to distribute the reactants to the active layers (electrodes: anode and cathode) respectively to collect and remove the products or inert gases from active layers. The gas distributor is often integrated into the bipolar plate. The gas diffusion layer is also called backing. Bipolar plates interconnect electrically a number of single PEFCs forming a so-called "stack" and multiplying thereby the voltage of the stack. An example of a PEFC stack

is given on the right-hand-side of fig. 2. The single cells and bipolar plates are covered on both sides with metallic end plates, which fix mechanically the stack and provide the inlets and outlets for the media. Near to each end plate one current collector (plus and minus pole of the stack) of the stack can be seen. It is obvious that within the stack manifolds are needed, which provide the media supply to each individual cell, whereby appropriate gaskets must separate the different media.

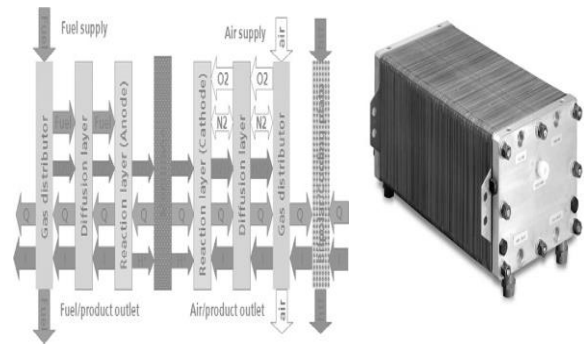


Figure 2: left: schematic representation of a PEFC. Right: PEFC stack, manufacturer NUVERA Fuel cells.

In summary the main transportation processes in the fuel cell are: (1) proton transport through the membrane from the anodic side to the catalyst surface of the cathode; (2) electrons through an external electric circuit from anodic side to the catalytic surface of the cathode (3) electrons from the cathode of one fuel cell to the anode of a second cell through the interconnecting bipolar plate, (4) the reactants and products to and from the reaction layers on the anodic and cathodic side as well; (5) heat from the membrane electrode assembly (MEA) to the htf cooling channels. There are various research activities ongoing regarding all components of fuel cells. In the following some aspects regarding material research on BPPs and polymer membranes are given.

IV. BIPOLAR PLATE AND MEMBRANE

Bipolar Plates (BPPs) contribute significantly to cost, volume and mass of fuel cell stacks. Hermann et al. report, that BPPs participate with about 80% to the stack weight and with 45% to its cost [5]. Newest research results at DLR-IFK indicate a contribution of about 33% to the stack cost [6]. A BPP fulfills multiple functions: it separates individual cells in the

- stack, it distributes fuel and oxidant, it may serve as a support for gaskets, it may serve as a cooling plate, it conducts electrons. Herman et al. give the following properties, which should be met by a BPP.
- ③ Electrical conductivity: plate resistance $< 0.01 \text{ Ohmcm}^2$
 - ③ Thermal conductivity: as high as possible.
 - ③ Hydrogen/gas permeability: $< 10^{-4} \text{ cm}^3/(\text{s cm}^2)$
 - ③ Corrosion resistance: corrosion rate $< 0.016 \text{ mA/cm}^2$
 - ③ Compressive strength: $> 22 \text{ lb/in}^2$
 - ③ Density: $< 5 \text{ g/cm}^3$

The following materials are under investigation for BPPs: non-porous graphite/electrographite; coated and non-coated metals; composite materials (polymer carbon and polymer-metal). Due to the character of the membrane, the BPP must withstand an acidic environment at temperatures around $80 \text{ }^\circ\text{C}$. Because of its chemical stability and low electrical resistance, graphite has been widely used in the past as preferable BPP material for PEFC stacks. However, the mechanical properties of graphite are not favorable. It must be handled with care, manufacturing of parts with structures is very expensive and the design of the entire stack has limits given by the mechanical properties of graphite. Therefore, metal and composite materials have actually drawn more attention.

Chemical stability is the main issue for metal plates, which can be shaped relatively easily. Recently the supplier Dana reported promising results for BPPs with special coatings [7]. In comparison with other BPPs the degradation of special coated plates was reduced about by a factor 6 compared to stainless steel. The observed degradation was even smaller than the degradation of a BPP made of gold.

As explained above main functions of the polymer electrolyte membrane are the conduction of protons from the anode to the cathode side of the fuel cell and the separation of the reactants. Consequently, the membrane should have high proton conductivity, low gas permeability, high thermal and chemical stability, and high mechanical stability. Perfluorinated ionomer membranes like Nafion® from DuPont are widely used in PEFCs. However, the automotive industry is asking for low-cost membranes, which can be operated at higher temperatures without the need for a sophisticated water management [8]. Worldwide

there are extensive research activities on advanced electrolyte membranes ongoing. An overview can be found in [9].

V. PRINCIPLES OF FUEL CELL

There are various types of FC systems. However, the principle of their function is similar. For a fuel cell system, three pillars are required: an anode, a cathode, and an electrolyte. FCs are categorized by the type of electrolyte material used. An FC can be composed of hundreds of individual cells, but each has the three same fundamental components. The electrolyte is located between the cathode and the anode. Figure 2 depicts a schematic of a polymer electrolyte FC (PEMFC) operation diagram. This FC type is also known as a proton exchange membrane FC. The PEMFC is what is most commonly used in mobile power applications, such as vehicles. While the electrolyte material used varies depending on the type of FC, the general function of the FC is as follows—fuel (pure hydrogen) is fed into the anode compartment of the fuel cell while air or pure oxygen is fed into the cathode side of the FC. On the anode side of the cell, electrons are separated as the gas tries to make its way through the electrolyte membrane. The membrane acts as a filter to separate the electrons and the hydrogen ions while only allowing the hydrogen ions to pass through. In the cathode compartment, the hydrogen ions that passed through the membrane combine with the oxygen atoms from the air supply to produce H_2O as a by-product; heat is also produced as a by-product. Unlike internal combustion engines, where the fuel is mixed with air and fuel, there is separation of the fuel and the oxidant with no combustion of the fuel in an FC. Therefore, FCs do not produce the harmful emissions that internal combustion engines produce.

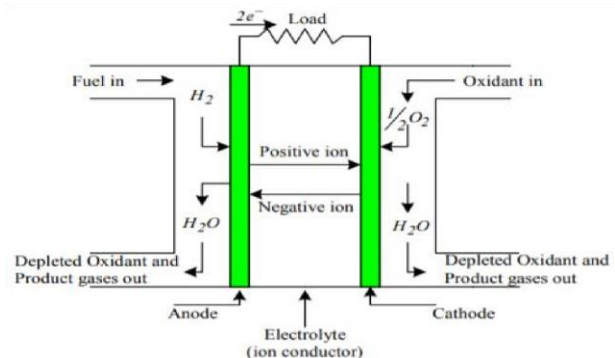


Figure 3. Fuel cell operation diagram.

VI. TYPES OF FUEL CELL

FC systems are classified by the type of membrane they use. Table 1 shows some of the more common FCs and the type of membrane each uses.

Fuel Cell	Abbreviation	Membrane
Solid Oxide Fuel cells	SOFC	Ytria-stabilized zirconia
Direct methanol fuel cell	DMFC	Solid polymer electrolyte (Nafion)
Phosphoric Acid fuel cell	PAFC	Phosphoric Acid (H ₃ PO ₄)
Polymer electrolyte fuel cell Or Proton exchange membrane	PEMFC	Solid polymer electrolyte (Nafion)
Alkaline fuel cell	AFC	Aqueous solution Potassium Hydroxide (KOH)

Table 1. Classification of fuel cell systems based on the employed membrane.

VII. UNITIZED REVERSIBLE FUEL CELL

A unitized reversible FC (URFC) is an energy-storage device that performs in water-electrolysis mode (EC mode) to produce hydrogen and works in FC mode to generate electricity. During the mode switching of the URFC, a reversible electrochemical reaction takes place that causes the change in temperature. Research on URFCs found pre-reactant switching, oxygen flow rate, hydrogen flow rate, and time interval length makes the URFC more efficient and reliable. The water accumulation in URFCs is a major problem, as it decreases the mass flow rate of reactants in FC mode. It also affects the mode switching in URFC. The residual water left during EC mode and the water produced during FC mode should be eliminated to facilitate smooth mode switching. Gas purging has

been used to remove the water at the proton exchange membrane at the end of the FC operation. Gas purge time increases with a decrease in the cell temperature. The gas purge flow rate should be greater than the purging time. Due to the high flow rates, the water droplets in the PEM can be pulled away from it. In FC mode, the mass flow rate of reactants is affected by the water content in the cell. To make the successful mode switching process from electrolysis mode to fuel cell mode, enough time should be provided. Allowing adequate time for gas purging enhances the mass flow rate, which helps the FC start up. The water in the channel is pushed by the oxygen gas, but the water in the oxygen side is still present due to the increased amount of time required for water electrolysis to occur. Pre-reactant switching is the method where the reactant gases are switched on before transitioning to the FC mode. By supplying the oxygen to the FC before the current supply, the residual water stored in the channels and the gas diffusion layer made during EC mode is eliminated. The gas must be supplied 180 s before the current transition to FC mode. This will effectively consume the residual water at end of EC mode.

VIII. FUEL CELL HYBRID VEHICLE

The development of one important aspect of the fuel cell vehicle—the electric motor—dates back to the early 19th century. Although electric vehicles were a strong contender in the early 20th century to become a mainstream transportation method, the ICE vehicle eventually won out due to the short range and the high cost of electric vehicles. Additionally, the discovery of Texan oil reduced the price of gasoline, thus it became affordable to the average consumer, which caused fuel cell and electric vehicles to take a backseat to the ICE vehicle for most of the last 100 years. The oil embargo in 1973 kick-started a renewed interest in FC power for personal transportation applications, as governments looked to mitigate their dependence on petroleum imports. In the early 1970s, K. Kordesch modified a sedan to operate from a 6-kW FC and a lead acid battery pack. The automobile was driven on public roads for about three years. In 1993, Ballard launched a fuel cell-powered light-duty transit bus using a 120-kW FC system, followed by a heavy-duty transit bus using a 200-kW FC system in 1995. In 1994 and 1995, H-Power built three fuel cell-battery hybrid buses, each using a 50-kW FC and a 100-kW nickel-

cadmium battery. The importance of these releases was to make FC technology understandable to key decision makers in industry and government. These buses helped to prove that fuel cells would work in the real world. Fleet-vehicle operations, such as buses and delivery services, were early adopters of fuel cell technology due to the ease of centralized refueling and the reduced requirement for a high range capability between fill-ups. Trials of FC powered buses have occurred in Vancouver and Chicago, as well as in other cities in Europe and North America. In July 2005, the first FC vehicle was leased to a family in California as an important step in getting more fuel cell vehicles on the road. However, many obstacles remain to be overcome before the FC vehicle can become a mainstream form of transportation. An obvious issue is the need for a hydrogen infrastructure to enable refueling of the vehicles. Hydrogen filling stations currently exist in many countries around the world, such as Canada, the USA, Iceland, Japan, Singapore, and Germany. Though these stations are currently not widespread enough to allow large numbers of people to begin driving FCVs, it is expected that more hydrogen infrastructure will be built as more FCVs become commercially available. Hydrogen FCVs have evolved significantly; currently, they can drive between 311 to 597 miles on a full tank. The development of these vehicles is increasing, but they still require significant improvements.

It should be noted that FCEVs are more promising in city bus applications due to two reasons—the supply of hydrogen is not crucial, because the buses refuel in one place, hence only one refilling point is required, and the price of FCs. FCs are still expensive therefore it makes more sense to buy them for vehicles that are in use for many hours each day. As of June 2018, there have been more than 6500 FCVs delivered to consumers. California was the leading market for FCVs, with nearly 3000 vehicles being delivered due to it having the largest network of hydrogen refueling stations (HRS). In Europe, Germany’s Linde AG and France’s Air Liquide have been working together to increase

Germany’s stations from 15 to 100 by 2017 and to 400 by 2023. About 1000 HRS would be required to provide full coverage in countries such as Germany or France, with a cost of 1.5 to 2 billion euro.

FCV sales volumes are projected to be significant, but only in the long term, even with a favorable climate-

policy scenario. Figure 4 shows FCV sales volume anticipations based on a long-term powertrain mix scenario (million annual units). Considering a similar scenario, the international energy agency (IEA) anticipates an FCV market share of about 17% by 2050 (35 million annual unit sales).

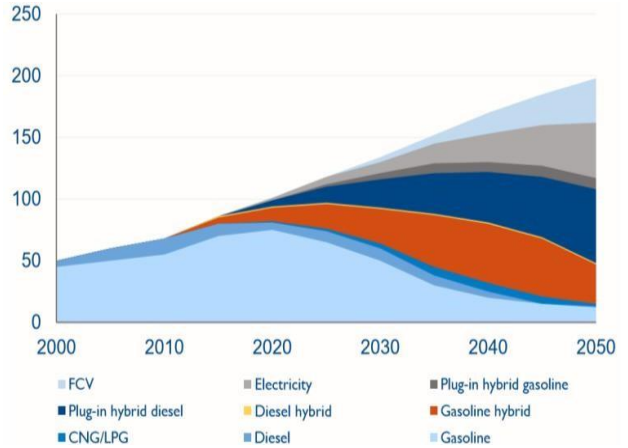


Figure 4. Fuel cell vehicle (FCV) sales volume.

Source: IEA. All rights reserved.

Another option to increase the HRS prevalence is to use electrolysis at refueling stations to convert electricity from the grid into hydrogen. This idea could be extended to residential applications, where people would have a hydrogen refueling station in their own homes. The obstacles associated with developing an adequate hydrogen infrastructure brings up another important question—where will the hydrogen come from? Although hydrogen is the most abundant element in the universe, it rarely exists alone in nature. Today, hydrogen is mostly produced by reforming natural gas. In this way, pollutants can be captured if the reforming is done at a central plant. However, other options, such as direct solar hydrogen from methane at landfills and hydrogen from bacteria, are continually being explored. If electrolysis is used to generate hydrogen, there may have to be an increase in electricity generation to satisfy the need for hydrogen, though the increase may be small, since hydrogen can be produced during off-peak times. Methods to generate electricity that have a minimal impact on the environment include nuclear, wind, solar, hydro, and geothermal. It is often said that, at the beginning of the hydrogen economy, most of the hydrogen will still be reformed from natural gas, but as time goes by, society will move towards more ideal sources of energy, such as wind and solar. Although

generation and transportation of hydrogen is a major issue in the deployment of fuel cell vehicles, many diverse ideas are being developed to solve the problem. Other obstacles for fuel cell vehicles include improving on-board hydrogen storage, improving fuel cell and battery durability, and increasing the efficiency and the performance of a fuel cell vehicle. The last barrier to commercialization is cost—a true challenge—as fuel cells are still extremely expensive today. Many parts used to work with fuel cells must be custom-made and can be very expensive as a result. However, these costs will decrease as technological processes are improved and components are mass-produced.

IX. CONCLUSION

Material research is a key factor pushing fuel cell vehicles forward. The paper gives some spot lights regarding material research on two main components of fuel cell vehicles – on-board hydrogen storage and fuel cell stack. The manifold functions of a PEFC stack require tailor-made materials, which can be mass-manufactured in order to meet low cost targets. The bipolar plate is an excellent example, where it is tried to replace graphite by metals or compound material. However, this requires answers regarding corrosion resistance and manufacturing processes. Looking on-board hydrogen storages, conventional storage technologies - compressed gas and liquid hydrogen - are approaching their limits, which are mainly defined by physical properties of hydrogen, when conventional solutions regarding construction principles and materials are applied. The new DLR concept for compressed gas storage could help to solve the cost and capacity issue together. However, it will be first applied to compressed natural gas vessels. Further extension of the concept to hydrogen vessels will be an extreme challenge. Promising results on complex metal hydrides are encouraging to intensify the material research in this field. However, basic research is still needed to identify material with sufficient high, reversible

storage capacities, having also a satisfying kinetics. In a next step storage in technical size could be developed.

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