Bipolar Plates and Gaskets: Different Materials and Processing Methods

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Isabel Kundler and Thorsten Hickmann

Abbreviations

BBP4	Phenolic resin bipolar plate (Brand name of Eisenhuth)
CNT	Carbon nanotubes
CR	Chloroprene rubber
EPDM	Ethylene propylene diene monomer
F	Fluor
FKM	Fluor caoutchouc material
H_3PO_4	Phosphoric acid
HNBR	Hydrogenated nitrile butadiene rubber
LCP	Liquid crystal polymer
LSR	Liquid silicon rubber
MFI	Mold flow index
MFR	Mold flow rate
NBR	Nitrile butadiene acrylonitrile rubber
NR	Natural rubber
PPS	Polyphenylene sulfide
PSU	Polysulfone
PVDF	Polyvinylidenfluoride
SBR	Styrene butadiene rubber
TGA	Thermogravimetric analysis
VMQ	Varmaq = Silicone-based solid raw material

19.1 Introduction

Fuel cells are basically galvanic cells converting chemical energy directly into electrical energy. Depending on the specific technology, this reaction may take place at elevated temperatures, thus technical requirements for materials and components of the stack become more and more challenging. The fuel cell technology has reached a considerable degree of technical maturity by now. However, market introduction still suffers from the challenge of supplying a high performing and reliable product on one hand and acceptable cost on the other hand. Besides the membrane electrode assembly (MEA), which is considered as the heart of a fuel cell, the bipolar plate is one of the key components of a fuel cell stack. Sizes and dimensions, weight, thermal and electrical properties of the stack are to a great extent determined by the bipolar plate technology.

In general, there are two different types of bipolar plate technologies: one based on metal and the other based on graphite as a conductive component. The latter can be pure (expanded) graphite or a graphite composite material containing a polymer binder. Metal plates offer excellent electrical and thermal conductivities and they can be machined to very small thicknesses. Additionally, they do not tend to form cracks or break even at small thicknesses and their nonporous structure offers good gas

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I. Kundler • T. Hickmann (✉)
Eisenhuth GmbH & Co. KG, Friedrich-Ebert-Straße 203, Osterode am Harz 37520, Germany
e-mail: t.hickmann@eisenhuth.de

Q. Li et al. (eds.), *High Temperature Polymer Electrolyte Membrane Fuel Cells*,
DOI 10.1007/978-3-319-17082-4_19

tightness. However, metal plates are inherently prone to corrosion under the aggressive oxidizing conditions in a fuel cell. Even high grade stainless steel does not resist the contact to both oxygen and electrochemical potential on the cathode side. Considerable effort has been devoted to the development of protective coatings for steel plates, both precious metal coatings and, preferentially, cost-effective coatings and surface treatments. However, for high temperature polymer electrolyte (HT-PEM) fuel cell applications the corrosion problem of metal plates appears to be unsolved by now and graphite-based materials are the preferred material according to current state-of-the-art.

The thermal and chemical conditions for materials applied in fuel cells are challenging, in particular in HT-PEM fuel cells. This fuel cell type operates at around 160 °C and in most cases in the presence of phosphoric acid in the electrolyte. As mentioned above, graphite composite-based bipolar plates with polymer binders are a superior material regarding their stability under corrosive conditions of acid contact and the presence of oxygen, electrochemical potential, and high temperature. However, composites require thicker plates than metal plates, resulting in more weight and volume of the stack. The MEA is always considered to be the dominating part of the stack in terms of cost and performance; however the bipolar plates have been underestimated both regarding their technical requirements and their contribution to the cost.

Graphite composite-based bipolar plates are manufactured using highly filled compounds [1]. They contain fillers like graphite or other electrically conductive carbons and binder polymers with appropriate heat deflection temperature. The key challenge is the competing interaction between electrical conductivity, provided by the carbon component, and mechanical stability as well as gas tightness, provided by the binder polymer. Both compounding and molding processes, which can be injection molding, compression molding, or others, are very sensitive to process parameters and need to be carefully controlled. The target is to manufacture bipolar plates in large volumes and with high quality, using standard methods for plastic processing such as injection molding. Only by using price attractive materials and the consequent focus on process automation when having higher volume, the bipolar plate can contribute significantly to a better market acceptance of fuel cells.

Besides the bipolar plate, the gasket material is an important component of the fuel cell stack and tends to be heavily underestimated. It plays a key role in the mechanical properties of the stack, compensated tolerances and partially determines the mechanical compression of the MEA. Inappropriately selected gasket materials may cause failure of the MEA or fracture of the bipolar plates. Despite the fact that the gasket has only to seal the stack, it is a highly challenging issue due to the tolerances of the other components which have to be managed. And last but not the least, the gasket has to be cost attractive.

Technically, the bipolar plate of the HT-PEM fuel cell stack has to accomplish the following functions [2, 3]:

- To conduct electrical current
- To conduct heat and distribute coolant in the cooling flow field
- To separate gases of adjacent cells
- To supply reactant gases homogeneously distributed in the flow field
- To transport product water (steam) out of the cell in the flow field
- To provide mechanical stability of the stack

The main functions of the gaskets in a HT-PEM fuel cell stack are

- To seal and prevent leakage of anode and cathode area
- To seal and prevent leakage of cooling plates
- Compensate tolerances and dimensional changes during temperature cycling
- Provide defined gap for the MEA or defined contact pressure between plate and gas diffusion layer (GDL) surface over a long time

Based on these basic functions of each component the technical requirements for plates and gaskets are described in the next section.

19.2 Technical Requirements of Bipolar Plates and Gaskets

Based on the technical functions described above, the US Department of Energy (DoE) suggested development targets for fuel cell components as shown in Table 19.1 for bipolar plates. These data are apparently based on communication with and input from conventional low temperature PEM fuel cell developers. Some of the data are directly transferred to the HT-PEM fuel cell technology. This has also been discussed by DoE [4] and Liao et al. [5]. However, depending on the specific application in HT-PEM fuel cells, priorities of the target values may significantly differ.

Additionally, the chemical resistance of the bipolar plate can be characterized by measuring the corrosion current under a potential typical for fuel cell operation and using 85 % phosphoric acid as an electrolyte. The relevance, detailed parameters, and development targets of this corrosion test are still subject to technical discussions and depend on the anticipated application of the plate. The accepted corrosion current under reference conditions is lower for long term co-generation of heat and power (CHP) applications than that for auxiliary power unit (APU) systems. The main functional requirements for the gaskets in the HT-PEM fuel cell technology are summarized in Table 19.2.

Additionally, the gasket material has to be resistant to the selected coolant under operating conditions. Since some rubber composites contain carbon fillers to improve their mechanical properties, unintended electrical conductivity of the material needs not be ruled out. It has to be noted that the values mentioned above are for orientation and refer to standard elastomer materials available on the market.

Based on these technical requirements, appropriate structural materials for both bipolar plates and gaskets have to be selected. In particular, the plate is exposed to challenging conditions since it faces high temperature, acid and oxygen contact, combined with an electrochemical potential. The plate has to provide mechanical integrity, gas tightness, and high conductivity over several thousand hours of operation, thus development and manufacturing of these materials is a challenging undertaking.

Table 19.1 Target specifications for bipolar plates in HT-PEM fuel cell applications

Technical property	Target value
Density	<2 g cm^{-3}
Electrical conductivity	>100 S cm^{-1}
Thermal conductivity	>10 W mK^{-1}
Flexural strength	>50 MPa
Shore hardness	>40
Temperature resistance	180 °C (thermo-mechanical test)
Phosphoric acid uptake	Low (specific target values depend on application and MEA technology)

Table 19.2 Proposed benchmarks for gaskets in HT-PEM fuel cell applications

Property	Target value
Density	<2 g cm^{-3}
Shore hardness	<70 Shore A, lower limit depends on application
Compression set	<18 %
Temperature resistance	180 °C
Chemical stability	Resistant against 75–95 % H_3PO_4 at 180 °C

19.3 General Concepts of Bipolar Plate Manufacturing

19.3.1 Compounding

As mentioned above, composite bipolar plates consist of a binder polymer which is highly filled with a conductive carbon component. Typical compositions are >80 % conductive filler and <20 % binder polymer. Compounding, processing, and manufacturing are substantially different from conventional polymers due to the high content of filler material in the compound [6]. The function of the carbon filler is to provide electrical and thermal conductivity. Therefore, a 3-dimensional percolating carbon structure is required. Usually, the main carbon component of the plate is synthetic graphite. Due to its crystalline layer structure the graphite platelets are inherently anisotropic in their physical properties. Electronic conductivity is provided by the mobility of electrons within the graphite layers of each platelet and not in transverse direction. Thus, for the bipolar plate the manufacturing process has to promote the formation of different orientations of the platelets to obtain isotropic physical properties of the macroscopic plate material. In the following, material aspects of bipolar plates are discussed in more detail.

19.3.2 Binder Polymers

In general, two different concepts of polymer binders can be applied in bipolar plates. First, the binder material can be polymerized or cross-linked in situ in the composite during molding of the plate (*resin method*). Second, a thermoplastic polymer material can be used in the compounding process (*thermoplastic method*).

In case of the *resin method* the graphite is carefully mixed with a prepolymer resin (e.g., phenolic resin) and a cross-linker and then compression molded to the final dimensions of the plate. During the molding process the cross-linking reaction is initiated thermally forming a 3-dimensional duromer network which is insoluble in any solvent under any conditions. The whole process needs to be carefully controlled in order to achieve the correct features of the flow field and to meet the tolerance requirements. The advantage of this method is a lower effort of compounding because of the rather low viscosity of the resin prepolymer. The disadvantage is that the polymerization and cross-linking process on the absorbing graphite process are rather difficult to control and may result in a broad distribution of chain segment lengths and inhomogeneous structures within the duromer network.

Eisenhuth's bipolar plates called BPP4 are developed for HT-PEM fuel cells and based on the phenolic resin technology. These plates are consistent and well performing for high temperature PEM fuel cells operating approximately 3000 h, e.g., in auxiliary power unit (APU) applications. However, for long term applications, such as combined heat and power generation (μCHP) with anticipated 20,000 h of operation, other materials may be preferred.

In case of the thermoplastic method a thermoplastic binder with a defined degree of polymerization is compounded with the graphite material. The chemical structure of the polymer remains unchanged during compounding or molding process. The polymer has to be selected with sufficient chemical and thermal stability (e.g., data from [7]). Several material candidates are available on the market in high quality and well-defined configurations. The disadvantage of the thermoplastic method for HT-PEM plates is the high effort in the compounding process which needs high temperature and strong mechanical forces. The same applies for the molding process, which requires a high amount of energy (heat and mechanical pressure). Both compression molding and injection molding are applicable for thermoplastic-based compounds. The main advantage of this method is a well-defined, reproducible composite material consistent in its chemical composition from lot to lot.

In both methods, after removal from the mold certain posttreatment procedures may be necessary, for example, to remove the skin of the mold release agent from the surface of the plate.

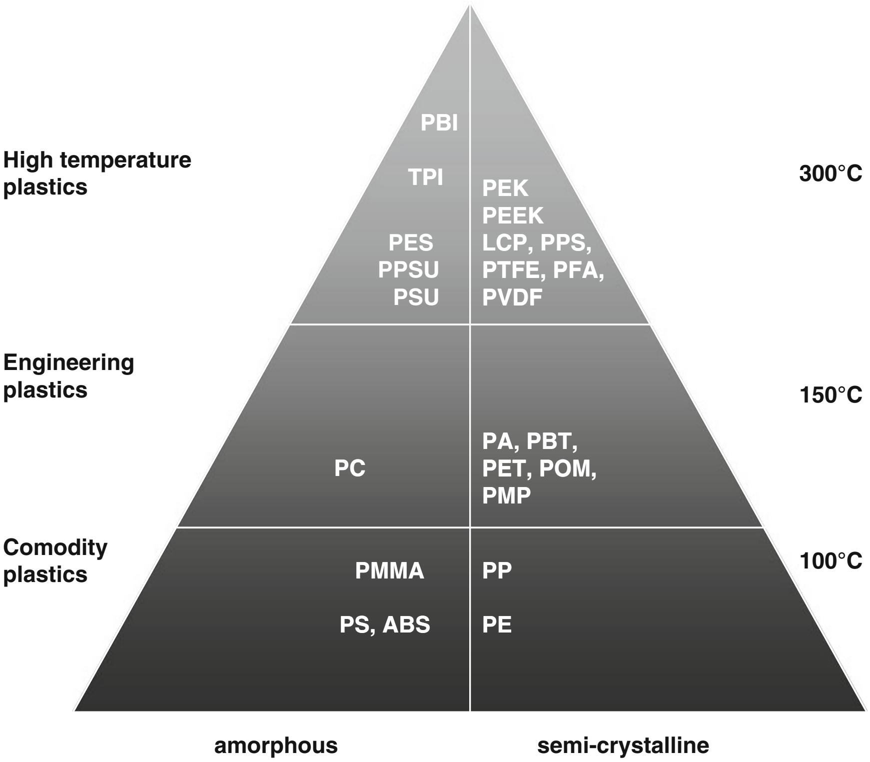
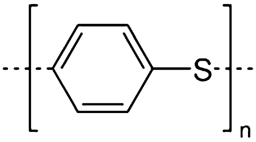


Fig. 19.1 The plastics pyramid according to thermal literature data of common polymers: preferred materials for HT-PEM applications are in the upper right region (*TPI* thermoplastic polyimide, *PES* polyether sulfone, *P* (*P*)*SU* poly(phenylene)sulfone, *PE*(*E*)*K* polyether(ether) ketone, *LCP* liquid crystal polymer, e.g., Vectra, *PPS* polyphenylene sulfide, *PTFE* polytetrafluoroethylene, e.g., Teflon®, *PFA* thermoplastic fluoro-copolymer, *PVDF* polyvinylidene fluoride, *PC* polycarbonate, *PA* polyamide, *PBT* polybutylene terephthalate, *PET* polyethylene terephthalate, *POM* polyoxymethylene, *PMP* polymethylpentene, *PMMA* acrylic glass, e.g., Plexiglas®, *PS* polystyrene, *ABS* acrylonitrile butadiene styrene copolymers, *PP* polypropylene, *PE* polyethylene)

Typical thermoplastic polymers as binders in HT-PEM bipolar plates are polyphenylensulfide (PPS), polyetheretherketone (PEEK), and derivatives of polyphenylsulfone (PSU). Looking at the plastics pyramid (Fig. 19.1), all of these polymers are in the upper right region of high temperature stability and semi-crystallinity. These polymers safely resist the acid and oxygen contact and provide heat deflection temperature far above the operating point of a high temperature PEM fuel cell.

Within the last years, considerable effort has been devoted to the development of innovative bipolar plate solutions at Eisenhuth. A broad variety of potential binder polymers have been tested. A major finding of the research activities is that semi-crystalline binders appear to be superior compared to amorphous polymers in terms of processability and mechanical properties of the plate material. Finally, the poly(p-phenylene sulfide) (PPS)-based material turned out to be the most well-suited polymer binder for HT-PEM fuel cell plates, so this chapter shall focus on PPS-based plates.

Fig. 19.2 Structure formula of poly (p-phenylene) sulfide (PPS)



PPS is a chemically and thermally highly stable material, consisting of aromatic rings linked with sulfur atoms (Fig. 19.2). It shows a glass transition temperature at 90 °C and a melting point at approximately 285 °C. The maximum long term service temperature (DIN53476) is 218 °C, thus significantly higher than the normal operating temperature of a HT-PEM fuel cell. PPS is an engineering thermoplastic polymer which can be molded, extruded, or machined to high tolerances.

Eisenhuth has developed and manufactured PPS-based composite plates and evaluated them both in situ and ex situ. In general, with PPS plates the same conductivity as known from the state-of-the-art and resin-based BPP4 material has been achieved (>100 S cm^{-1}, meeting the targets of DoE). The detailed test data are to be discussed below.

19.3.3 Graphite Materials and Fillers

Carbon, most commonly graphite-based, provides at least 80 wt % and even more in volume percent of the bipolar plate material. While synthetic graphite materials are the basis of bipolar plates, several carbon additives can be employed in order to boost conductivity properties of the composite material. Examples for such additives are highly conductive carbon nanotubes (CNT) or high surface area carbon blacks. Carbon blacks can be formed by thermal decomposition of hydrocarbons in the gas phase [8] and a broad variety of different materials with different surface area, hydrophobicity, and conductivity are available. It has to be considered that high surface area carbons are more prone to undesired corrosion effects than graphite-based materials, thus their positive conductivity effect has to be balanced against long term stability requirements. This proposes to use conductivity boosters rather for auxiliary power unit (APU) and uninterrupted power supply (UPS) applications and employ stable graphite materials for long term CHP usage.

Another important aspect of carbon materials is purity. Since most fuel cell membranes and catalysts are highly sensitive to contamination with Fe-ions and other metal residuals, the raw materials for bipolar plates have to be carefully characterized with respect to their Fe content. The carbon or graphite type also mainly determines the properties of the bipolar plate like porosity, phosphoric acid uptake, and hydrophobicity, both regarding the surface and the bulk. For the HT-PEM fuel cell technology, preferred configurations are hydrophobic structures with low phosphoric acid uptake.

One of the key properties of the bipolar plate is electronic conductivity. This is generated by a 3-dimensional percolating network of conductive graphite or carbon particles. The graphite/binder system is always inhomogeneous and can be considered as a two-phase system of conductive carbon paths and a polymer matrix for mechanical integrity. Figure 19.3 shows the scanning electron microscopic (SEM) images of a bipolar plate compound, where the crystalline and amorphous regions in the graphite are visible. The mesoscopic binder polymer distribution cannot be characterized with REM because of similar average electron densities of carbon and the polymers. The mesoscopic structure of the material highly depends not only on the chemical composition, but also on processing, such as compounding and molding parameters. The overall process from the raw material to the molded plate has to be carefully controlled to ensure consistency and reproducibility of the bipolar plates from lot to lot.

19.4 Characterization Data of HT-PEM Bipolar Plate Materials

19.4.1 General Remarks

Certainly, the final success criterion for any bipolar plate is the in situ performance and stability under real fuel cell operating conditions. However, fuel cells are highly complex systems with numerous sources of inconsistency. Thus, ex situ bipolar plate characterization is required for material development and quality control. Several test methods are well established for bipolar plates and a few selected shall be presented here with an example for test results.

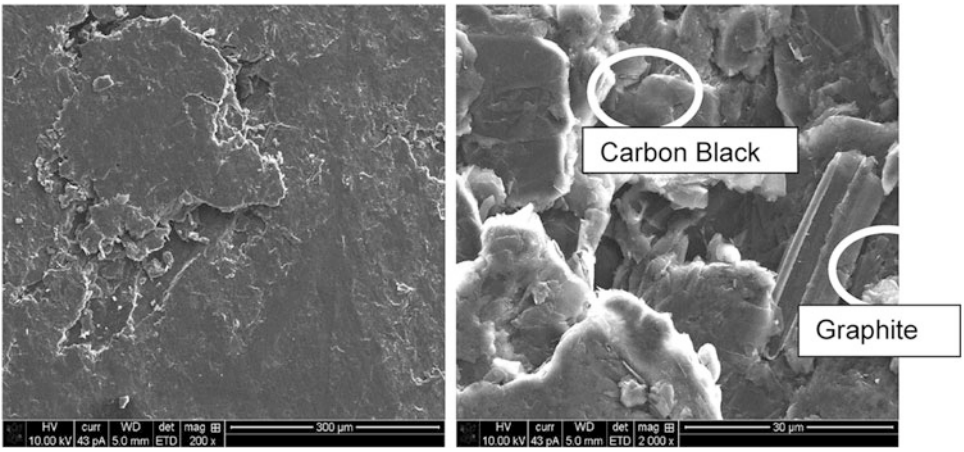


Fig. 19.3 Scanning electron microscopy (SEM) of a bipolar plate compound

19.4.2 Thermogravimetric Analysis

During fuel cell operation HT-PEM bipolar plates are exposed to high temperatures up to about 180 °C and sometimes even higher. Consequently, all raw materials used for plate manufacturing have to be resistant to approximately 200 °C, assuming 20 °C as a safety distance for a prolonged period.

Figure 19.4a shows examples for thermogravimetric analysis (TGA) analysis for state-of-the-art HT-PEM plates of type "BPP4" and low temperature LT-PEM materials as a reference. Recently developed HT-PEM materials with high temperature resistant thermoplastic binder polymers (PPS and PSU Ultrason®) are shown in Fig. 19.4b.

As expected, the LT-PEM material clearly failed at temperatures below 200 °C showing a significant decrease in weight due to chemical degradation of the binder polymer. The TGA of phenolic resin-based BPP4 material shows a slight decrease in weight when approaching the 200 °C benchmark and then continuously degrades between 200 and 400 °C. This can be explained by a broad distribution of chain segment lengths of the duromer network and residual oligomers from the resin because the polymer structure has been generated on the graphite surface without any purification steps after polymerization or cross-linking.

In contrast, the two bipolar plate samples containing thermoplastic binders (here PPS and PSU Ultrason® from BASF) show a constant weight up to >400 °C, followed by a well-defined weight drop at >500 °C, far beyond the relevant temperature range for PEM fuel cells. The results clearly show the benefits of thermoplastic high temperature resistant binders compared to in situ polymerized resin-based materials: the more defined structure of thermoplastics enables better reproducibility of the composites and excellent thermal stability over the full temperature range of HT-PEM operation. Under real fuel cell conditions, e.g., operating at 170 °C, BPP4 was successfully tested for systems with approximately 3000 h of operation. However, for long term CHP applications, e.g., with a target 20,000 h, the bipolar plate material needs to be improved. In this case, the PPS-based bipolar plate, currently under development by Eisenhuth and research partners, is a candidate providing improved thermal and mechanical stability.

Not only the binder polymer, but also the carbon fillers have to be characterized with respect to their thermal stability. In the next TGA example, several PPS-bonded bipolar plate samples with different contents of highly conductive carbon nanotubes (0–4 % CNT) are investigated by TGA. As shown in Fig. 19.5, the peak of thermal decomposition at approximately

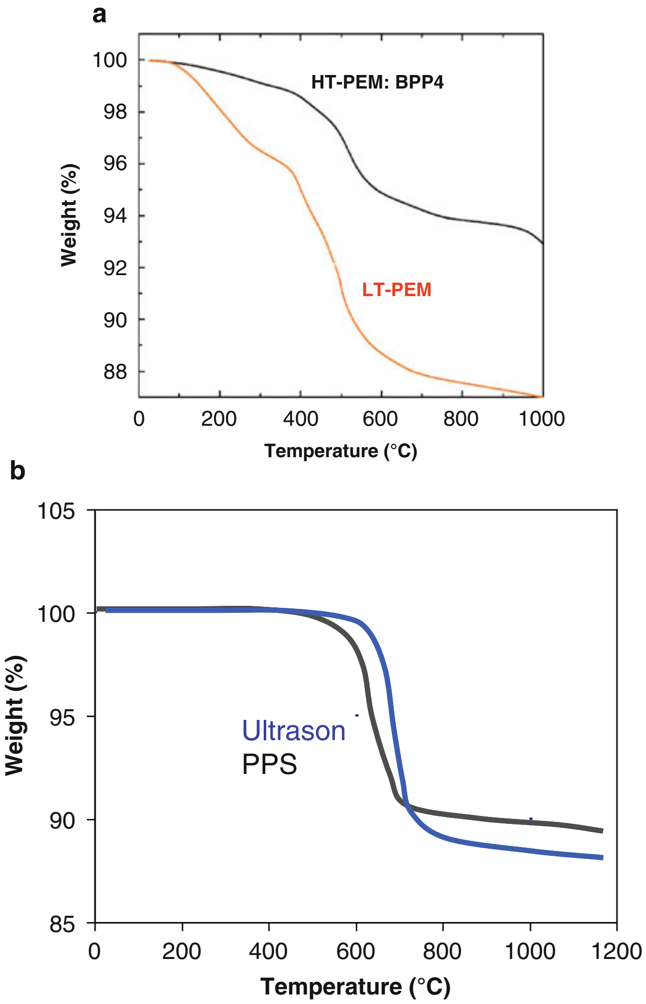


Fig. 19.4 Thermogravimetric analysis of state-of-the-art HT-PEM material BPP4 compared to LT-PEM bipolar plate (**a**) and the advanced HT-PEM bipolar plates based on high temperature polymer binders of Ultrason and PPS (**b**), heating rate 10 K min^{-1}, inert atmosphere [9]

550 °C is equal for all samples. However, in the region between 200 and 300 °C a small weight loss was detected, which seems to be related to the CNT concentration. Despite this reaction takes place at temperatures higher than the HT-PEM fuel cell, it has to be considered as a potentially significant degradation mode for long term operations, e.g., in CHP applications. Further experiments with conductivity boosters will be performed in the near future in order to evaluate the critical limits for these additives. These results are preliminary and further experiments are necessary for conclusions about critical parameters and relevant degradation mechanisms from carbon additives.

19.4.3 Electrical Conductivity Measurements (In-Plane)

Clearly, electrical conductivity both in-plane and through-plane is one of the most important properties of the bipolar plate. Despite most fuel cell (component) laboratories have access to electrical conductivity testing equipment, by now there is no standardized test method for

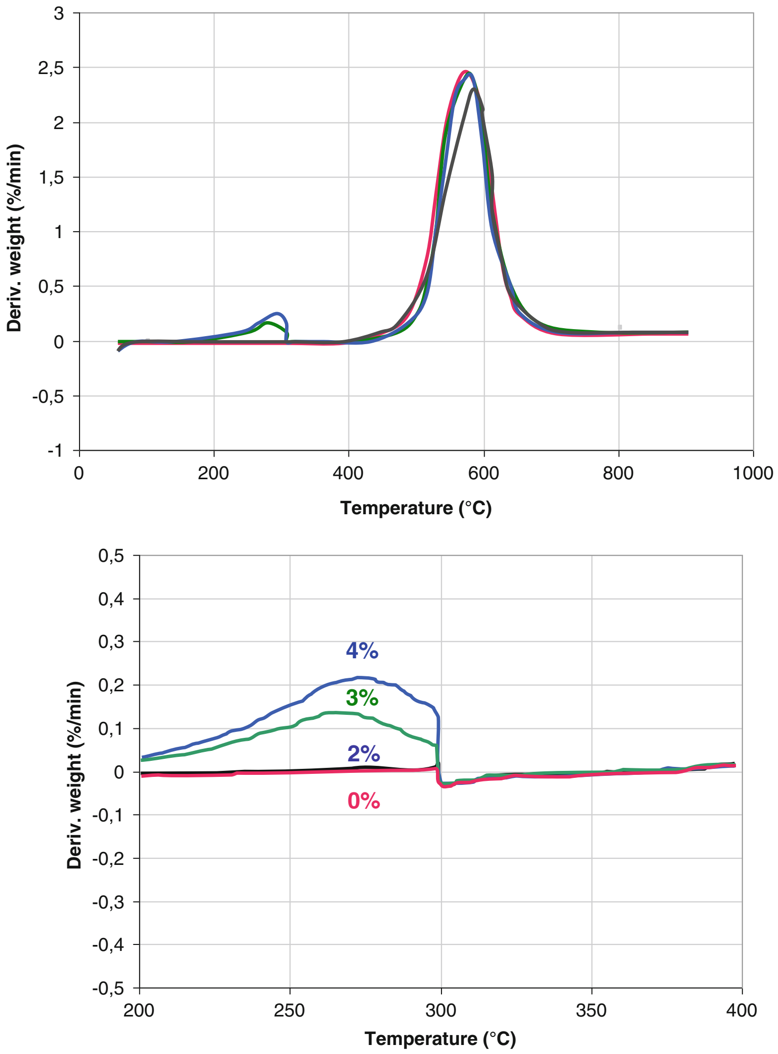


Fig. 19.5 Thermogravimetric analysis of PPS-based HT-PEM plates doped with 0–4 % carbon nanotubes CNT

bipolar plates, and comparing results from different sources can show significant differences even if the same samples are tested. One of the main reasons may be surface effects and pre-treatment of the sample. As shown in Fig. 19.6, Eisenhuth has implemented an own testing system for this application, which is suited for in-plane conductivity testing.

The in-plane conductivity device allows for a conductivity mapping over the whole sample area of 200 × 300 mm^2. Therefore, it can characterize the material with respect to the degree of homogeneity. Conductivity mapping is an important tool both for quality control and for material and process development. For graphite composite plates it is well known that compounding and molding are highly sensitive to process details and may generate inhomogeneous structures of surface and bulk of the material. Certainly, the development target is a homogeneous distribution of conductivity with only minimal deviations between different points on the sample. In BPP4 plates the compounding and manufacturing process are mature and well controlled, and the conductivity mapping shows an even distribution with values of

Fig. 19.6 Testing device for in-plane electrical conductivity measurement at Eisenhuth

approximately 100 S cm^{-1} (0.01 Ω cm). In case of the newly developed PPS materials the manufacturing process is under development and some material samples still show a significant relief structure in the conductivity mapping. Two examples of the mapping for PPS-based plates are shown in Fig. 19.7.

In the case of the (a) sample the data propose the molding process as a source of inconsistency because the same pattern—higher resistivity on the lower and right edge—was found in several samples. One explanation for these results is that graphite platelets are oriented parallel to the surface when filling the mold and therefore less contribute to conductivity than in the case of a fully isotropic material.

19.4.4 Electrical Conductivity Measurements (Through-Plane)

Theoretically, in an isotropic material in-plane and through-plane conductivities are supposed to be equal. However, in many bipolar plate materials the through-plane values tend to be lower than their in-plane counterparts. One explanation for this effect is that during the molding process graphite platelets are oriented preferentially parallel to the sample's surface, thus transporting the electrons faster within the in-plane direction than the through-plane. In Fig. 19.8 examples for through-plane results are presented, with the key messages that (a) the through-plane conductivity tends to be systematically lower than in-plane (approximately 0.02 instead of 0.01 Ω cm in-plane) and (b) in mature materials, such as resin-based BPP4, the values covers smaller range due to better control of the process. In development materials, such as PPS of different configurations and fluoropolymer-bonded PVDF types, the range is broader and small changes of process parameters still cause significant deviations in conductivity. In the course of process and product development for each new material this will be improved.

19.4.5 Phosphoric Acid Uptake of Bipolar Plates

HT-PEM fuel cells operate with phosphoric acid doped polymer membrane as electrolyte. The acid is physically adsorbed to the membrane. The phosphoric acid distribution within the fuel cell components, such as membrane, catalyst layers, microporous layer, gas diffusion layers, and bipolar plates, is known to be a critical parameter for performance and life time of this type of fuel cells [10]. There are no defined specifications about phosphoric acid uptake of the bipolar plate because its impact on the fuel cell performance strongly depends on several parameters and always has to be considered in a context of the overall fuel cell design.

In general, the phosphoric acid electrolyte is required to conduct protons in the membrane and a certain limited amount of the doping acid is needed to provide ionic conductivity in the catalyst layers. In the GDL (microporous layer and bulk) as well as in the bipolar plate, phosphoric acid is not required and even highly undesired because it compromises the gas transport properties in the GDL and may catalyze

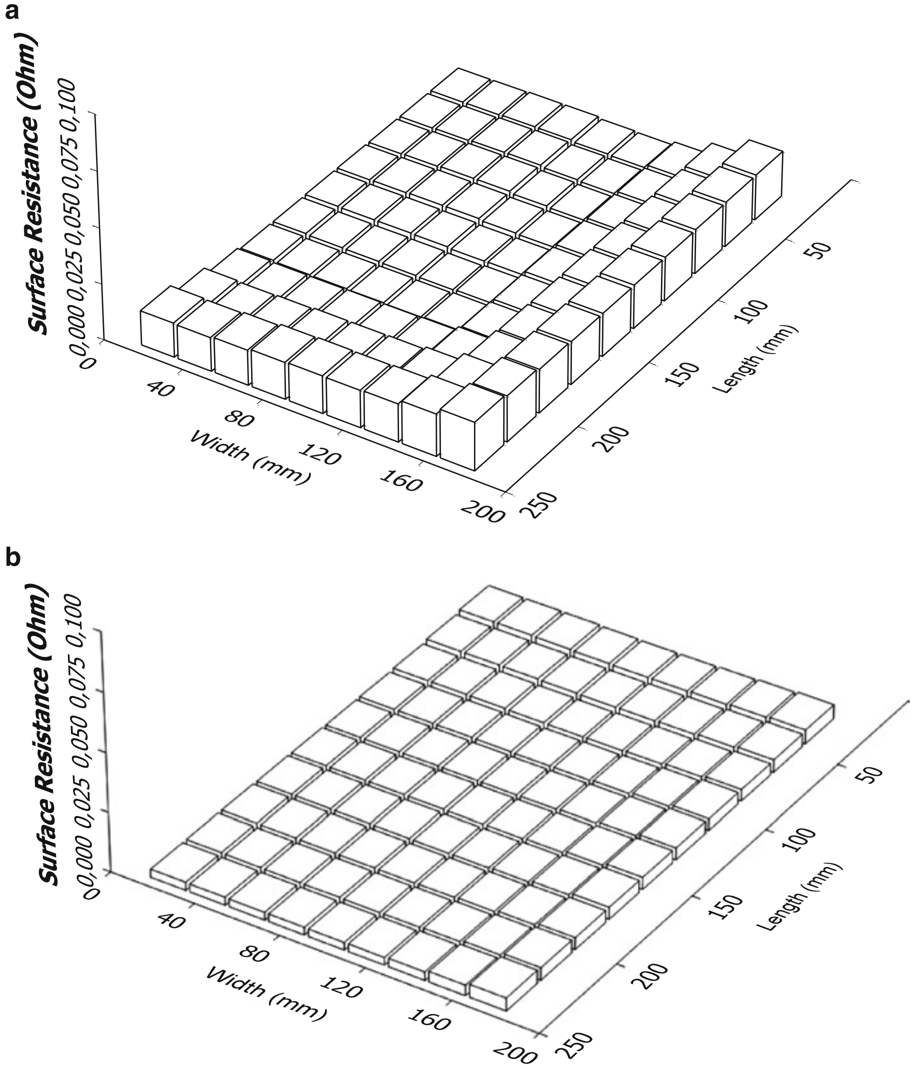


Fig. 19.7 Surface resistance mapping (in Ohm, relative test method the characterize homogeneity) of two high temperature PEM bipolar plates. (**a**) The material shows a higher resistance at the edges with 80 % of data points between 0.01 and 0.02 Ω (**b**) The material displays a lower resistance and improved homogeneity, all data points <0.01 Ω with only minor deviations

undesired oxidation processes in the bipolar plates material. Any phosphoric acid uptake of the bipolar plate can have the following effects:

- The bipolar plate may compete with the membrane and catalyst layer regarding the acid retention. Then degradation of the cell performance occurs because of insufficient ionic conductivity in one of these components, although the plate itself does not become damaged by the acid.
- Acid contact accelerates carbon corrosion in all carbon components of the cathode. However, the high surface area and Pt-loaded carbons in the catalyst layer are significantly more prone to this degradation mechanism than the graphite of the bipolar plate. Nevertheless, this is considered and the plates will

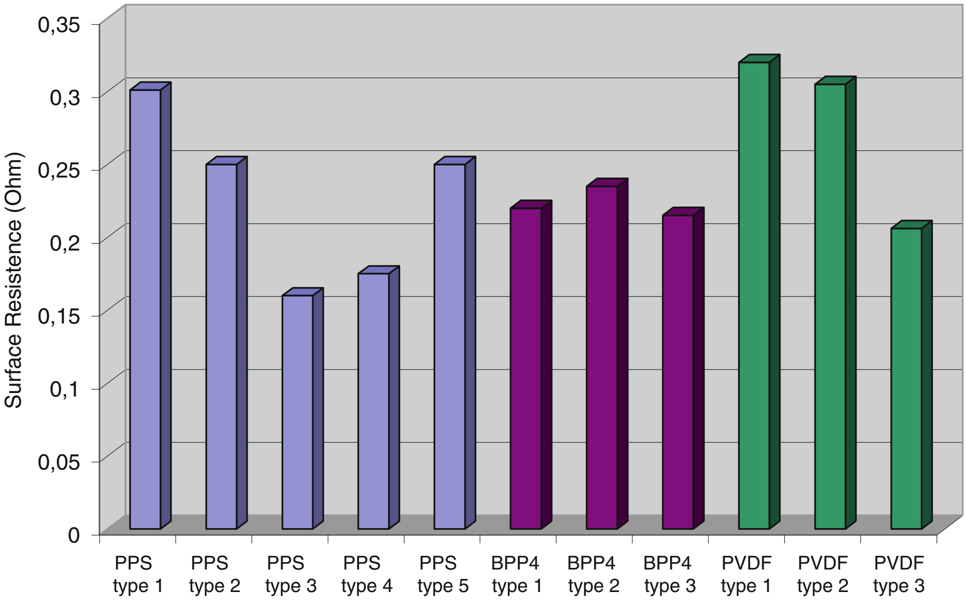


Fig. 19.8 Through-plane surface resistance (relative test method) results of HT-PEM bipolar plates with different configurations where BPP4 is mature material while PPS and PVDF are development materials. The data show higher homogeneity of the mature BPP4 material compared to the new configurations with PPS and PVDF (Status as of 2014)

be further improved with respect to their corrosion stability—within the frame given by thermodynamics.

- A certain acid concentration in the bipolar plates causes a vapor pressure of phosphoric acid in the gas stream and may prevent against acid evaporation in the catalyst layers and membrane. In this case, the acid content in the plate may have a positive effect on long term stability in certain applications.

All these arguments are only for a brief overview, show the complexity of the system and cannot at all be considered as a complete set of information or even as a recommendation for certain membrane, MEA or bipolar plate configurations.

Eisenhuth has characterized different bipolar plates with respect to their acid uptake. In general, a low acid uptake, corresponding to a high hydrophobicity, is desired in order to keep the acid management in the overall cell under control. The acid uptake is mainly determined by the carbon component (graphite), which constitutes >80% of the volume of the material. However, also the binder may influence the porosity in particular on the surface of the plate. Detailed studies about structure–property relationships regarding acid uptake with different materials are in progress at Eisenhuth. Preliminary results are shown in Fig. 19.9. Here, different bipolar plates have been tested in an HT-PEM rainbow stack for more than 3000 h. After disassembling, the plates were characterized by titration and the phosphoric acid uptake in a unit of gram per cm^2 plate (or MEA respectively) was calculated. While the two samples PPS and PVDF, both containing the same synthetic graphite and a different binder polymer, show very similar values and, in general, a low acid uptake, the plates containing other carbon materials strongly absorbed phosphoric acid. This material was manufactured using another carbon type, yielding a completely

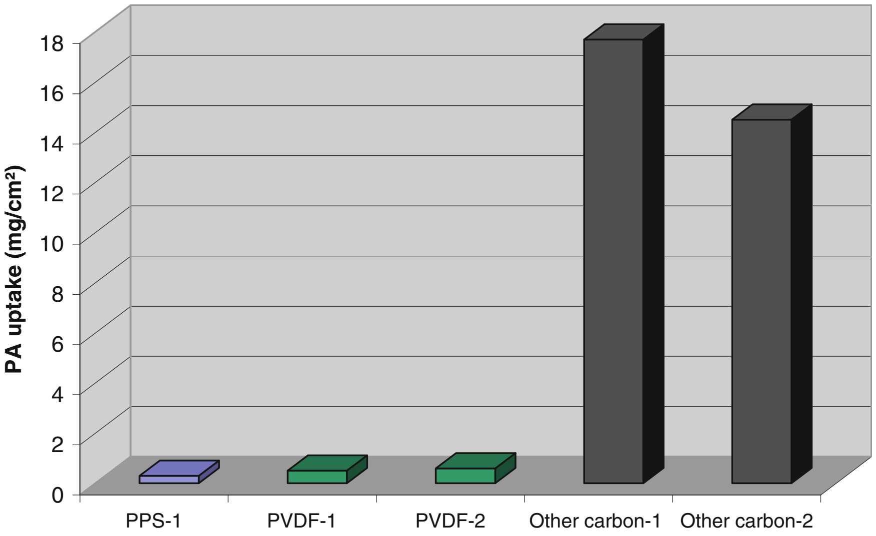


Fig. 19.9 Phosphoric acid uptake in different bipolar plates after >3000 h HT-PEM fuel cell stack operation. The samples based on PPS and PVDF contained the same type of synthetic graphite, whereas the sample called other carbon contained another type of carbon. It shows the strong contribution of the carbon, and only minor contribution of the binder polymer (test data kindly provided by NEXT ENERGY, Oldenburg, Germany)

different behavior with respect to hydrophobicity and porosity.

19.5 Gaskets

The fuel cell developers have for many years invested tremendous efforts in improvement and technological readiness of the core components, such as membranes, electrodes, GDLs, the overall MEA configuration, and also the bipolar plates. The significance of gasket materials has been underestimated until the last years. Despite the gasket does not directly contribute to the electrochemical processes, inappropriate gaskets can cause leakage or coolant contamination of the MEA. Additionally, in particular for phosphoric acid containing fuel cells, gasket thinning can cause MEA over-compression, resulting in over-compression of the membrane causing gas crossover through the membrane. On the other hand, too thick gaskets can cause MEA under-compression, resulting in a high contact resistance between GDL and plate. These examples clearly indicate that gasket properties and stability significantly contributes to life time and reliability of fuel cells. The key functions of the gaskets are:

- To seal the cell
- To catch up tolerances (from membrane, GDL, plate, and gasket)
- To provide appropriate compression of the MEA over a long time

In case of the HT-PEM fuel cell technology the compression of the MEA or the membrane respectively is known to be a critical parameter for performance and stability. The gasket thickness has to be appropriately selected, also considering potential creep of the materials. Hard gaskets support well-defined gaps, however may be compromised in their sealing properties, do not compensate tolerances very well, and may cause mechanical distortion on the bipolar plates, which in turn can cause cracks or beak after a long time. On the other hand, with soft gaskets it is more difficult to control the compression ratio, but they are superior with respect to tolerances and mechanical stabilization of the stack. Beside the mechanical properties, thermal and chemical stability as well as compatibility with a

Table 19.3 Gasket material overview with physical properties

Description/intern. term	NR	SBR	EPDM	CR	NBR	HNBR	Si/VMQ	FKM
Hardness shore A[a]	35–95	40–95	25–85	30–90	20–95	40–90	25–85	50–90
Tear strength N mm^{-2}	30	25	20	29	25	25	8	20
Elongation at break %	800	450	450	450	500	500	300	400
Temperature min./max.	−0.63	−0.5	−0.5	−0.32	−0.29	−0.2	−0.35	−0.09
Steam resistance	0	++	++	++	+	++	++	++
Oil resistance	−	−	−	+	++	++	+	++
Acid resistance	+	+	++	+	0	+	−	++

[a] Hardness measured with a shore durometer according to standard ASTM D2240, an established standard test method for polymers and rubbers

nonaqueous coolant are crucial material criteria of the gaskets. In general, like the other HT-PEM fuel cell components the gaskets have to resist temperatures up to 200 °C, concentrated phosphoric acid contact and oxygen contact. Fluoroelastomers (FKM) are most likely the material of choice for HT-PEM fuel cell applications, and for certain applications the ethylene propylene diene monomer (EPDM) rubber may be a cost efficient alternative. The arguments clearly show that gaskets are a highly customized component for each stack manufacturers. For an overview, some typical gasket properties for a broad variety of materials are listed in Table 19.3.

In order to supply consistent gaskets with appropriate tolerances the viscoelastic properties of the gasket prepolymers are an important parameter. A low viscosity is beneficial for processing. For plastic materials usually the mold flow rate (MFR) or mold flow index (MFI) are specified by the supplier, supporting the manufacturer for plastic parts with information relevant for processing properties. However, these data are standard data and not always compatible with the molding conditions or equipment at the part manufacturer. In addition, for rubber materials or their prepolymers these data are not available in most cases. Therefore, Eisenhuth developed a phenomenological test method to characterize polymer materials with respect to processability, as schematically represented in Fig. 19.10. In this test, a polymer is pressed into a helix-shaped mold with a defined pressure under process-relevant temperatures. The viscous polymer flows into the helix mold and finally stops, when the applied pressure is equal to the “back-pressure” of the mold. The length of the helix can be correlated to the low viscosity or good processability. This mold can also be used to find the processing windows of materials as far as the viscosity is concerned.

Figure 19.11 presents a typical result of the helix test. As mentioned above, the length of the extended helix is a measure of the processability. This test has been performed with a variety of potential gasket materials to achieve a data baseline. The values are listed in Table 19.4. The results show that the processability depends on the type (silicon > fluoro-elastomer FKM) and also on specific material configuration (FKM “low viscosity” > regular FKM).

Fig. 19.10 Helix mold for elastomer processability characterization (by Eisenhuth)

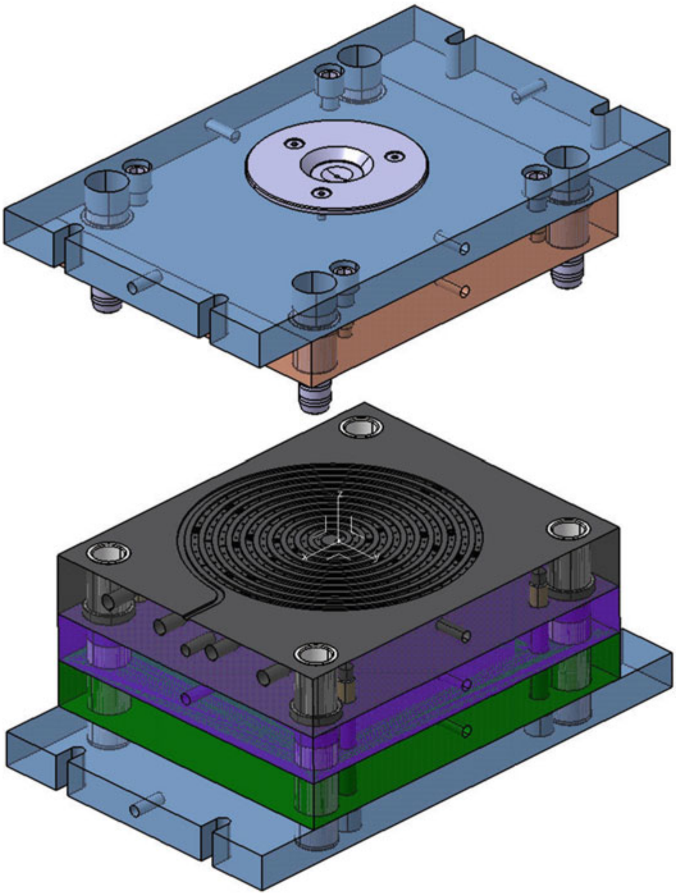


Fig. 19.11 Elastomer sample after vulcanization in the helix test

Table 19.4 Results of the helix test for potential HT-PEM gasket materials (in the Eisenhuth mould)

Material	Length (cm)
EPDM 60 shore A	37.9
EPDM 40 shore A	41.0
EPDM 50 shore A	47.0
FKM 60 shore A—low viscosity	109
FKM 65 shore A	59.5
FKM 47 shore A	14.5
Silicon, 30 shore A	123.4
Silicon, 45 shore A	104.4
Silicon, 60 shore A	124.5

Acknowledgements The authors acknowledge fruitful collaboration with, extensive test work by, and the positive relationship with TU Clausthal, Next Energy Research Institute of University of Oldenburg, ZBT (Research Center of Fuel Cell Technology) in Duisburg. Thanks are also extended to the colleagues from Elcore and Truma for their input and inspiration. Public funding is gratefully acknowledged from BMWi/Projekttraeger Juelich in the project “Rationalisierung” (#0327849), “HT-Dicht” (03ET2050A) and from the European Commission in the project CISTEM (325262).

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