A review of the research progress of composite bipolar plates for proton exchange membrane fuel cells with different substrate materials

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Abstract. As an important component in proton exchange membrane fuel cells, the performance of bipolar plates, such as electrical conductivity, bending strength, corrosion resistance, gas tightness, etc., directly determines the lifetime of fuel cells and their promotion applications. Composite bipolar plates have become a research hotspot and have been applied and promoted in related fields because of their strong design, flexible formulation, excellent performance, low cost, and applicability to various operating environments. This paper analyzes in detail the research progress of composite bipolar plates using different matrix materials, briefly analyzes and introduces the research on improving the performance of composite bipolar plates, and makes an outlook on the future development trend of composite bipolar plates for proton exchange membrane batteries.

Keywords: Proton exchange membrane fuel cell; Composite bipolar plate; Single-phase system matrix composite bipolar plate; Multi-phase matrix system bipolar plate.

1. Introduction

Since the twenty-first century, with the rapid socio-economic development, the economies relying on traditional fossil energy sources are consuming more and more, while the total amount of traditional fossil energy sources is decreasing. On the environmental side, the combustion of fossil fuels also produces large amounts of polluting emissions such as NOx , SOx , etc., which makes the world is facing a serious energy shortage and environmental pollution problem [1]. At the same time, at the level of energy utilization, because most of the traditional energy utilization relies on the Carnot heat engine for energy conversion, the energy utilization efficiency of even the best jet engine is only about 60% due to the limitations of the Carnot cycle, and the widely used internal combustion engine has an energy utilization efficiency of only about 40%, and most of the energy is wasted [2]. This makes the large-scale sustainable use of traditional energy sources a serious challenge. It is widely understood that improving energy utilization and developing clean new energy sources are the most convenient ways to solve this problem [4,5].

Among many new energy sources, secondary energy sources such as hydrogen and methanol are regarded as clean and efficient, safe and sustainable, and economic alternatives to fossil fuels, and are the most promising clean energy sources and strategic energy development direction for human beings in the 21st century. Fuel Cell (FC) is one of the important applications of secondary energy, and as an economic alternative to fossil fuels, it has great potential for large-scale commercial production and social application. Fuel cell is a power generation device that converts the chemical energy in fuel directly into electricity through electrochemical reaction. Classified by electrolyte, fuel cells generally include proton exchange membrane fuel cells, phosphoric acid fuel cells, alkaline fuel cells, solid oxide fuel cells and molten carbonate fuel cells. Due to the modular design of fuel cells, they can be scaled up to suit the needs in various settings [1-5].

The proton exchange membrane fuel cell (PEMFC) is a class of fuel cells, which is also known as polymer electrolyte membrane fuel cell [6]. It is an energy conversion device that can efficiently utilize clean energy sources such as hydrogen, with energy conversion rates as high as 40% to 60, and even up to 90% if waste heat is fully recycled, and causes little environmental pollution. Proton exchange membrane fuel cells usually use perfluorosulfonic acid type ion exchange membrane as electrolyte, Pt/C as electrocatalyst, air or oxygen as oxidant, and graphite or surface modified metal plates with gas flow channels as bipolar plates, thus converting chemical energy stored in the fuel and oxidant directly into electrical energy. PEMFC is characterized by high energy conversion efficiency, environmental friendliness, fast start-up at room temperature, etc. It is suitable for a variety of applications such as transportation, power station, portable power and submarine, and has a broad market prospect. It has attracted the attention of more and more countries and enterprises, which have invested heavily in this project.

The bipolar plate (BP) is one of the core components of a proton exchange membrane fuel cell, accounting for 80% of the mass and about 30% of the cost of a PEMFC stack [7].The wide application of PEMFC requires bipolar plates with, among other characteristics. The main role of the bipolar plate is to deliver the reaction gas to the membrane electrode through the flow field on the surface, while collecting and conducting the current and discharging the heat of the reaction and the reaction product water. Therefore, the bipolar plate must have certain properties: good gas tightness, good mechanical properties, low cost, good conductivity to ensure the electrical connection between the monomers, suitable flow field channels for the reaction gas as well as the coolant, good corrosion resistance, good heat dissipation and the ability to discharge the reactants (water) in time [8].

The materials used to prepare PEMFC bipolar plates are mainly graphite, metal and composite materials. Graphite bipolar plate has the advantages of good corrosion resistance, electrochemical stability, good electrical conductivity and good thermal conductivity, however, graphite material its microstructure is low in bending strength and porous and prone to fracture due to the atoms between layers bonded by van der Waals forces with large spacing. In addition, the high cost and long cycle processing method of graphite sheet is not suitable for mass production. Metallic materials with good mechanical properties, high electrical conductivity, high gas tightness, good dense and gas barrier properties, low raw material cost and easy processing have also gradually become popular materials for bipolar plate preparation in recent years [10]. However, metal bipolar plates are prone to corrosion and passivation, and the precipitation of metal ions may cause catalyst poisoning in fuel cell membrane electrode assemblies (MEAs), while the passivation layer produced by metal bipolar plates can increase the surface contact resistance and affect the service life of PEMFC stacks. These problems can be partially solved by surface modification, but this keeps the processing cost of the material high. The composite bipolar plate is a kind of bipolar plate made of graphite, carbon black, carbon nanotubes, carbon fiber and other conductive fillers, and non-conductive materials such as epoxy resin as the substrate through hot pressing, injection molding and other processes, which overcomes the poor mechanical properties of graphite bipolar plate, processing difficulties and poor corrosion resistance of metal bipolar plate and other challenges, while having the advantages of low cost and resistance to use required for mass production. It is now a key direction of bipolar plate material research [11]. Although the conductivity and thermal conductivity of composite bipolar plates are slightly lower than those of pure graphite and metallic bipolar plates, the performance of the prepared bipolar plates can still meet or even far exceed the requirements of PEMFC by optimizing the composite composition, ratio and forming process.

Table 1. U.S. Department of Energy (DOE) performance targets for bipolar plates from 2020 to 2025

Characteristic	Unit	2020 Status	2025 Target
Electrical conductivity	S/cm	≥100	≥100
Flexural strength	MPa	≥25	≥40
Anodic corrosion current density	$\mu A/cm^2$	<1 and no active peak	<1 and no active peak
Cathodic corrosion current density	$\mu A/cm^2$	<0.1	<1
Area Ratio Resistance	Ω-cm^2	<0.01	<0.01
Gas permeability coefficient (80°C/3atm)	Std- cm^3 /scm^2 -Pa	$<1.3\times10^{-14}$	2×10^{-6}

Quality	kg/kW_{net}	0.4	0.18
Cost	USD/kW_{net}	3	2
Molding elongation	%	40	40

According to the classification of matrix materials, composite bipolar plates include single-phase matrix composite bipolar plates and multi-phase matrix system bipolar plates. The single-phase matrix system composite bipolar plates have been studied the most among the carbon-based polymer materials and are now widely used in commercial production. Multi-phase matrix system materials present better electrical properties due to the continuity of the internal structure, which is promising to be the most widely used bipolar plate for PEMFC applications and is the key direction for the research and application of PEMFC composite bipolar plates [12]. So far, the main single-phase matrix materials used to prepare composite bipolar plates are thermosetting materials, thermoplastic materials, and materials with inorganic salt cement as the matrix. Multi-phase matrix composites are prepared by choosing two or more non-conductive raw materials consisting of non-combined materials as matrix, mainly carbon black particles or carbon nanotubes as conductive fillers. In this paper, a systematic review of the current research progress of composite bipolar plates and the future development trend of bipolar plates is presented mainly around the matrix materials of bipolar plates.

2. Single-phase matrix system composite bipolar plate

Composite bipolar plates are made using dispersed conductive fillers such as fibers, powders, and flakes mixed with a continuous nonconductive matrix, and most of the research is aimed at improving the mechanical properties and reducing the cost of graphite bipolar plates. The electrical properties of the bipolar plates are provided by the fillers, while the mechanical properties are provided by the non-conductive matrix [12]. The electrical properties in composites increase with the filler content, but the mechanical properties of the material decrease due to the decrease in matrix content. Therefore, how to obtain the best balance between electrical and mechanical properties is the focus of research on composite bipolar plates with unidirectional matrix systems [14].

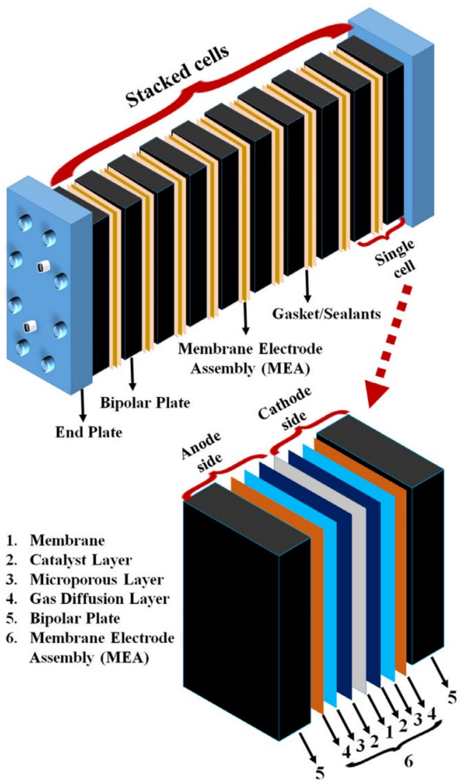

Figure 1.Schematic diagram of the main components and stacking of the PEMFC single cell [61]

The conventional composite bipolar plate has a single non-conductive substrate, which is generally selected from thermosetting materials such as epoxy resin and phenolic resin, thermoplastic materials such as polyethylene (PP) and polyvinylidene fluoride (PVDF), and inorganic salt materials such as silicate cement. These materials replace the expensive graphite in conventional graphite plates, and therefore can significantly reduce the production cost of pole plates. The matrix in the composite improves the bending resistance and processing performance of the pole plate, while alleviating the problem of excessive permeability in the porous graphite material. The organic matrix exhibits better corrosion resistance than metallic bipolar plates in the acidic environment during PEMFC operation, and eliminates the surface treatment process necessary for metallic bipolar plates in terms of processability, thus enabling the production of composite bipolar plates with higher robustness, including thinner thicknesses and finer flow fields, through a more efficient production process than expensive and slow machining [15].

In addition, composite bipolar plates are characterized by the ability to choose different combinations of conductive fillers and substrates and different raw material ratios to meet the requirements of different application conditions of fuel cells, thus making composite bipolar plates more flexible in terms of production applications [16].

2.1 Epoxy resin-based bipolar plates

Epoxy resin (EP) is the most mature and widely used thermosetting polymer. It has the advantages of good mechanical and thermal properties, low curing shrinkage, chemical resistance, good heat dissipation, high flexural strength, and excellent processability [17]. Conventional epoxy resin bipolar plates generally use natural graphite in the form of powder, flakes or fibers with high content (50-80%) as conductive filler, the morphology and properties of graphite are determined by the mining, powdering and filtering processes and generally have high electrical conductivity [18]. However, in the high price due to the decreasing reserves of high-quality natural graphite year by year, artificial graphite can also be taken as a filler, but artificial graphite needs to be prepared by heat treatment of carbon powder or compounds at very high temperatures (2500-3000°C), this material may not be fully graphitized, and natural graphite has better thermal conductivity and anisotropy, so the performance of artificial graphite is slightly worse than natural graphite [19]. In recent years, different conductive fillers that can be used in epoxy composites have been discovered, such as carbon nanotubes (CNT), carbon fibers (CF), etc. On the one hand, these materials are less costly than high-quality natural graphite. On the other hand, materials such as carbon nanotubes, carbon fibers, graphene (Gr) and their derivatives are reliable reinforcing materials for EP-based composites due to their inherent excellent mechanical, thermal and electrical properties [20].

From the current research literature, with the development of advanced material preparation processes, more current literature in the application of epoxy resin composites for the preparation of bipolar plates take multi-walled/single-walled carbon nanotubes, graphene, carbon fibers as conductive fillers, molded by molding process, and supplemented by surface treatment, addition of materials, improved processing methods and other means to seek the best ratio of ingredients, processing technology to improve their electrical properties and mechanical properties [21-29].

The more typical epoxy resin matrix bipolar plates such as the composite plates developed by GrafTech AET are already widely used in commercial production. The GRAFCELL® used to prepare the plates is a proven thermoset composite material consisting of a high content of expanded or exfoliated natural graphite and an epoxy resin. The bipolar plates made from this material have the advantages of high electrical conductivity, low contact resistance, high corrosion resistance, low gas permeability, high thermal conductivity, 33 times higher thermal diffusion coefficient than stainless steel, easy processing by molding, and low cost. However, the low bending strength of this bipolar plate is one of the main disadvantages of this bipolar plate due to the high graphite content. This leads to a larger thickness of the pole plate, which makes the bulk power density increase. The low bending strength greatly affects the operational stability and durability of the bipolar plate due to the inhomogeneity of the reaction gas in the battery stack and the high pressure drop operating environment [21].

Current research considers the use of nanotubes, carbon fibers (CF), and other materials with higher aspect ratios as fillers as a significant advancement in epoxy composite systems compared to the previous use of graphite and carbon black particles (CB) due to the over-permeability theory. epoxy composites with mass fractions of carbon nanotubes ranging from 0.001 to 1 wt % and compared the electrical conductivity with the osmosis threshold of carbon black particle-epoxy composites. The results showed that MWCNT composites formed a good conductive network inside the composite and the over-permeability threshold decreased by several orders of magnitude, which allowed the material to achieve higher conductivity than conventional epoxy composites without degrading the mechanical properties [22].Joong Hee Lee et al. investigated the effect of carbon filler types including graphite, carbon black, multi-walled carbon nanotubes, and carbon fibers on composite bipolar plates prepared from powder-type epoxy resin compression. molding on the performance of composite bipolar plates prepared by powder-type epoxy resin. It was concluded that with the gradual addition of fiber conductive fillers such as MWNT and CF, the conductivity and flexural properties of the composites increased rapidly. On the contrary, although particulate fillers such as carbon black and graphite can improve the conductivity of the composites, the bending properties of the pole plates deteriorate significantly at higher loads due to the higher content. The electrical conductivity and bending properties of the bipolar plates were significantly improved with the incorporation of small amounts of MWCNT and CF composites compared to graphite/epoxy composites [23].

Jong Wan Kim et al [24] improved the mechanical properties and in-plane conductivity of conventional composite bipolar plates by inserting several layers of continuous carbon fiber fabric (CFF) into graphite powder (GP)/epoxy composites. The bipolar plates prepared with this material had good stability at 300 °C. The composite bipolar plates containing 70-75% carbon filler had the highest in-plane conductivity of >180 S/cm, which was much higher than the target set by DOE, and their flexural strength was highest after the insertion of five layers of CFF, reaching 131.2 MPa. The results indicate that the in-plane conductivity of continuous fiber composites can be overcome by introducing the GP / CFF hybrid system. The poor formability and lower in-plane conductivity of the bipolar plate and the weaker bending performance of the GP composite, while GP can compensate for the lower conductivity of the CFF material through the plane, forming a benign complement.

It was partially found [19] that the mixing of graphite and carbon nanotubes produced a coordination effect that inhibited the aggregation of the filler in the matrix, which led to a lower threshold of overpermeability of the filler, resulting in a good conductive network and improved electrical conductivity and flexural strength.Majid Niaz Akhtar et al [18] used multi-walled carbon nanotubes, natural graphite (NG) and epoxy resin by compression molding The composite bipolar plates were fabricated by compression molding technique using multi-walled carbon nanotubes, natural graphite (NG) and epoxy resin. The resulting bipolar plates had a conductivity of 129 S/cm, a conductivity of 16 S/cm through the plane, a flexural strength of 38 MPa, a Young's modulus of 17 GPa, and the highest Shore hardness of its kind in bipolar plates with a value of 70.

Kyungmun Kang et al [25] proposed a multilayer BP structure, an ultrathin multilayer bipolar plate consisting of a combination of carbon fiber prepreg, pure graphite layer and graphite-epoxy resin composite layer and prepared by a molding process. The thickness of the multilayer prepreg BP is about 0.6 mm thick with good electrical behavior (in-plane conductivity of 172S/cm and through-plane conductivity of 38S/cm) and a high-quality serpentine flow channel structure. This study greatly improves the low through-plane conductivity and poor plasticity of the original prepreg. This bipolar plate can greatly reduce the volume of the electric stack, however the extremely low thickness of this bipolar plate may lead to a lower mechanical strength, which may limit its application scope.

Fatih Darıcık et al [26] optimized the modification ratio of carbon nanotubes to prevent overutilization of the material and achieve BP with higher cost advantage.The best performance was achieved with the addition of 1.25% CNT and the electrical conductivity of the bipolar plate reached

120 S/cm and other properties were similar to that of aluminum alloy (AA 3105) bipolar plate. The bending strength and bending modulus were increased by 42% and 27% to 47 MPa and 33 GPa, respectively, compared with the BP without the addition of carbon nanotubes. the DOE targets were met.

Some of the studies found that doping metals in composites can effectively reduce the resistivity of bipolar plates.Fatih Darıcık et al [27] designed a mono-type bypass aluminum/composite hybrid bipolar plate with low resistance in order to reduce the resistivity of carbon fiber/epoxy composite bipolar plates. The researchers fabricated a composite bipolar plate with low resistivity and low permeability after surface modification of the aluminum sheet and carbon fiber/epoxy composite by mechanical grinding technique in order to obtain the maximum contact area between the aluminum sheet and carbon fiber, and finally, the surface of the developed bipolar plate was modified by electromagnetic-carbon modification technique to remove the excess resin and contaminated impurities on the surface. Jun Woo Lim et al [28] developed a monotype hybrid bipolar plate and aluminum foil for PEM fuel cells to reduce the energy loss due to the resistance of the PEM fuel cell system components. The surface of the aluminum foil was sanded with 320 grit sandpaper, preformed into a runner shape, covidally bonded to the surface of a carbon fiber/epoxy composite, and coated with a thin graphite flake to form a soft layer that increases the contact area with the gas diffusion layer (GDL). An extended aluminum bypass is bonded to the edge of the GDL to finally form a continuous structure of GDL - bipolar plate - bipolar plate - GDL. The resistivity of this bipolar plate was shown to be only 2% of that of the conventional bipolar version.Min-Chien Hsiao et al [29] prepared a new metal mesh hybrid polymer composite bipolar plate for PEMFC by inserting a copper or aluminum mesh into the vinyl ester/MWCNT composite. This bipolar plate reached a maximum electrical conductivity of 643S/cm and a maximum thermal conductivity of 30.4 W/m-K . Such ultra-low resistance bipolar plates with metal addition can be used in high power fuel cell systems.

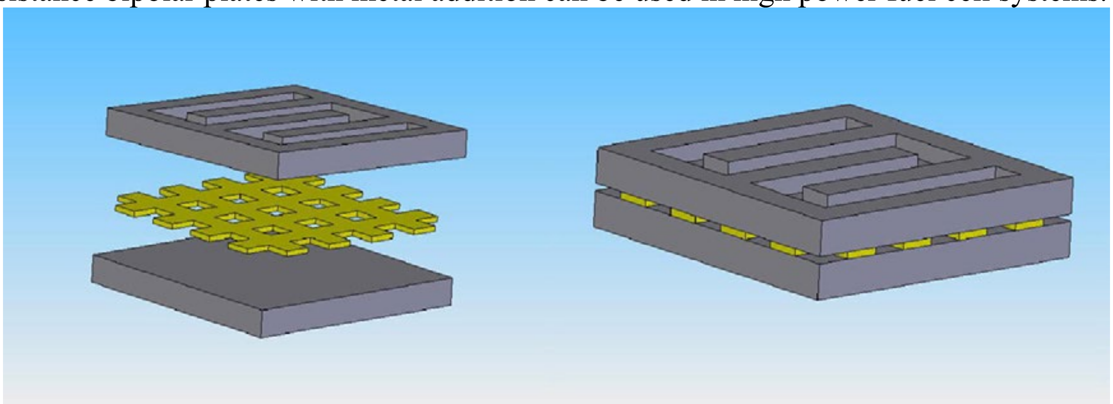

Figure 2. schematic diagram of the structure of the metal mesh hybrid polymer composite bipolar plate [81]

In addition, there is a lot of literature focusing on the search for new improved epoxy resin bipolar plates from bipolar plate structure, surface modification, coating, and improved processing [30-39].

Most current fuel cell bipolar plates require the use of additional rubber gaskets to seal the cell stack, which requires additional curing processes at high temperatures and increases fabrication and assembly time, which is extremely detrimental to commercial production.Jun Woo Lim et al [30] developed an innovative gasketless carbon fiber/epoxy composite bipolar plate that was able to without the use of additional gaskets or curing cycles achieve high gas tightness, and this process greatly saves assembly time and processing cost. At the same time, the bipolar plate achieved a planar resistivity of 0.02 Ω-cm^2 at a compaction pressure of 2 MPa, which is close to the target value specified by DOE.

Carbon fiber-reinforced epoxy resin bipolar plates have the potential to form resin-rich areas during processing, which allows the carbon fibers to be buried by the resin and thus increases the area specific resistance (ASR) of the bipolar plate. -The "soft layer method" solved the above problem by using a thin polymer release film between the compression mold and the composite material. Ultimately, the area specific resistance was reduced to 18 mΩ/cm^2 , meeting the 30 mΩ/cm^2 target established by the U.S. Department of Energy. Although the tensile strength of the planar specimens was reduced by 5% due to the increase in stress concentration, the flexural strength of the channel specimens with channels made by the soft-layer method was increased by 12% because the soft layer covered the rough surface of the mold. In addition, the soft-layer method did not affect the gas permeability, which remained at zero during the 100-h test period, but the soft-layer method still had the drawback of excessive processing pressure. Researchers developed a nonwoven carbon felt/epoxy resin composite bipolar plate by the soft-layer method in 2017 and improved the soft-layer method [32]. The bipolar plate with the best overall performance was obtained at 1.38 MPa with an area specific resistance of 20 mΩ /cm^2 , Young's modulus and tensile strength of 25.1 GPa and 79.3 MPa, respectively.Bu Gi Kim [33] proposed a selective surface treatment technique developed using electromagnetic field and carbon black (electromagnetic-carbon surface treatment). This method was able to remove the non-conductive resin from the surface without damaging the carbon fibers, giving them a low resistance. It was found that the surface-treated composite bipolar plates had a lower resistance than conventional composite bipolar plates, with a total resistance of only 0.45 Ω at a processing pressure of 1 MPa.This electromagnetic-carbon surface treatment can be applied to the production of composite bipolar plates quickly and efficiently.Ha Na Yu et al [34] developed a plasma etching treatment to reduce the contact between GDL and continuous carbon/epoxy composite bipolar plates resistance. The longitudinal and transverse tensile strengths of the bipolar plates reached 750 MPa and 400 MPa, and the area specific resistance was reduced to nearly 13 mΩ/cm^2 .

The coating method is also an important means to reduce the contact resistance. Ha Na Yu et al [35] coated a thin layer of graphite on a conventional carbon/epoxy bipolar plate to reduce the contact resistance between the bipolar plate and the gas permeation layer. When the graphite coating thickness was 50 μm, the graphite coating on the carbon/epoxy composite bipolar plate accounted for 10% and 4% of the total electrical resistance and interfacial contact resistance of the conventional carbon/epoxy composite bipolar plate, respectively.Minkook Kim et al [36] developed a new bipolar plate using a pure woven carbon fiber/epoxy composite combined with the graphite coating method. Compared with the composite bipolar plate using continuous carbon fiber prepreg, it showed an improvement in electrical conductivity and fabrication productivity in the cross-thickness direction. A graphite layer was coated on the surface of the plain carbon/epoxy composite plate to reduce the interfacial contact resistance between the GDL and the bipolar plate, while the graphite coating achieved near-zero gas permeability. The bipolar plate was experimentally measured to have a flexural strength of 110 MPa and an area specific resistance of 25 mΩ/cm2 , both of which met the standards set by the U.S. Department of Energy.

Chao Du et al [37] from Dalian Institute of Chemical Physics conducted an optimization study on the preparation process of compressed expanded graphite/epoxy bipolar plates. The authors discussed three different preparation techniques: compression-impregnation, impregnation-compression, and compression-impregnation-compression methods, and concluded that all three methods can obtain bipolar plates that meet DOE requirements, among which the compression-impregnation-compression method is the best method to produce the material properties when the resin content is 25%. sang One Kim et al [38] used a hot compression curing process in Prior to the hot compression curing process, a resin extrusion process was used to remove the excess resin and increase the volume fraction of carbon fibers by inducing proper resin flow thereby increasing the electrical conductivity of carbon fiber/epoxy bipolar plates. The volume fraction of carbon fibers in the prepared slotted BP increased with increasing resin extrusion pretreatment temperature, and the ASR of the slotted BP decreased from 17.5 mΩ-cm2 to 7.6 mΩ-cm2 , a reduction of 56%, after resin extrusion pretreatment at 120°C. Fabrizio Roncaglia [39] investigated the adhesion of the substrate and conductive filler and developed a Fabrizio Roncaglia [39] studied the adhesion of substrates and conductive fillers, developing a "wet mixing" method with the addition of isophorone diamine (IPDA) curing agent during the mixing of raw materials and optimizing the production process parameters. The average values of conductivity and flexural strength of the bipolar plates prepared from the obtained materials were 162S/cm and 48 MPa, respectively.

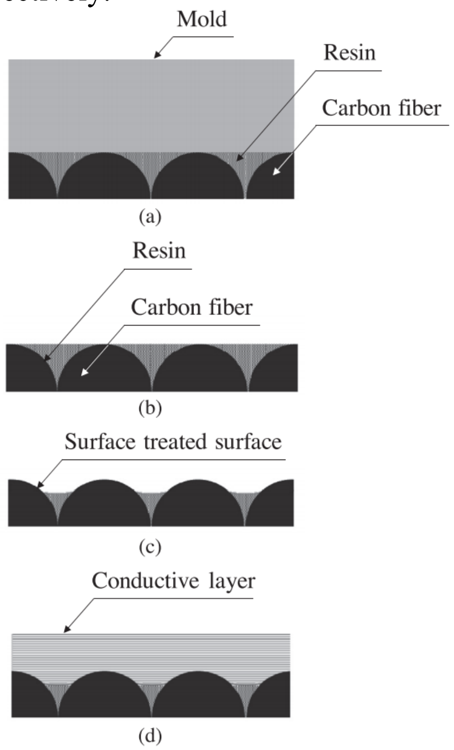

Figure 3. surface morphology of the bipolar plate surface after treatment with different processes: (a) compressive forming process; (b) no treatment; (c) surface treatment; (d) coated coating [31].

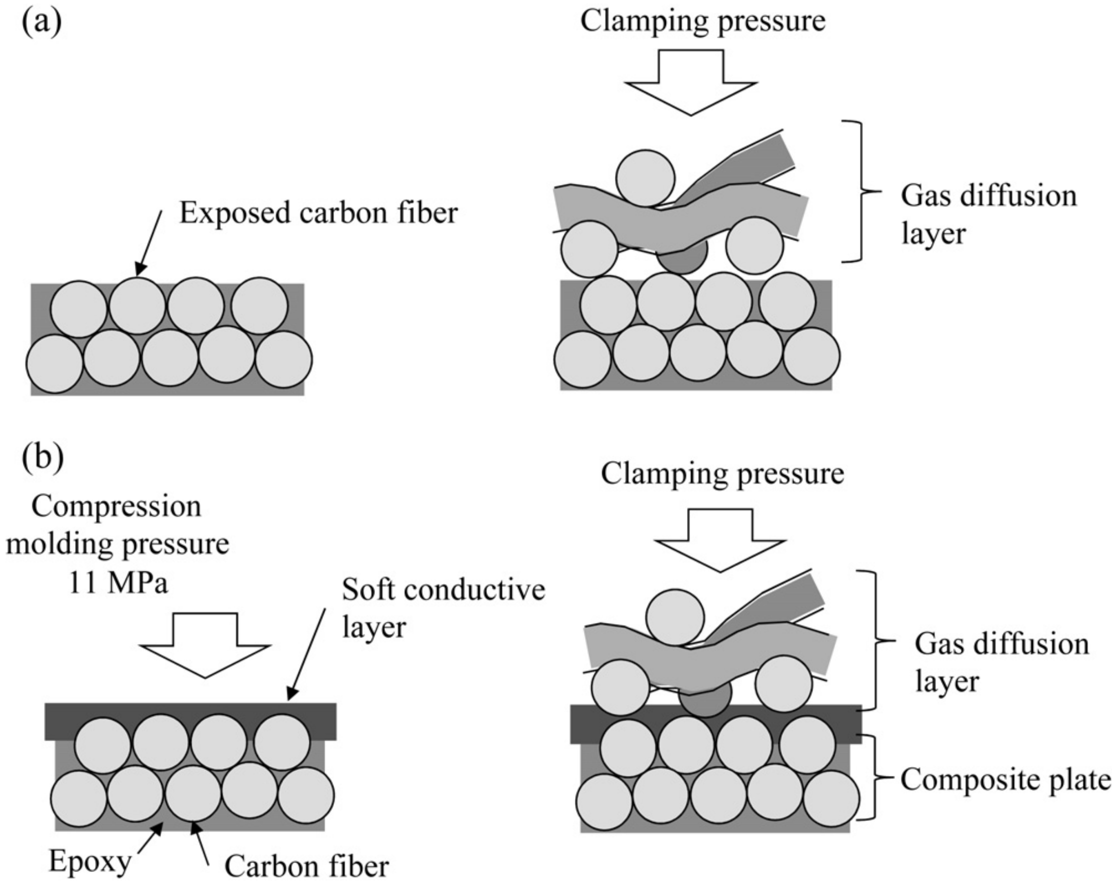

Figure 4. schematic diagram of surface treatment methods for bipolar plates: (a) mechanical grinding method; (b) graphite coating method [15]

2.2 Phenolic resin-based bipolar plates

Phenolic resins such as phenol-formaldehyde based resins have been developed for more than 100 years and have good acid resistance, mechanical properties, and heat resistance, and are now indispensable polymeric materials for electronics, machinery, construction, aerospace, military, and defense [40]. Compared with epoxy resins, phenolic resins have better heat resistance and have a faster curing rate than epoxy resins at certain temperatures and have a cheaper cost. However, phenolic resins require heating and pressure during curing; have water as a byproduct of the reaction, so there are bubble residues and have the disadvantage of large curing volume shrinkage; and have poorer alkali resistance than epoxy resins; and their flexural strength is lower than epoxy resins [41]. In the application of composite bipolar plates, similar to epoxy resin matrix materials, phenolic resin-based bipolar plates are prepared from high content of conductive fillers, such as graphite, carbon black, etc., mixed with phenolic resin by hot pressing process. Due to the good heat resistance, short curing time and lower price of phenolic resin, recent literature has focused on the development of high temperature bipolar plates based on phenolic resin composites and the development of processes for rapid mass production of bipolar plates [42].

Typical phenolic resin-based bipolar plates have a wider selection of conductive fillers, probably due to the higher curing temperature compared to epoxy resins, which results in a more uniform dispersion of the fillers.

Kyungmun Kang et al [43] investigated the effect of different graphite fillers on the electrical conductivity and bending strength of composite plates. The flake NG-based bipolar plates exhibited the most superior electrical and mechanical properties due to the high crystallinity and aspect ratio of natural flake graphite (NG).

Liu Hongbo et al [44] from Hunan University prepared phenolic resin-based bipolar plates with phenolic resin as the binder, different types of graphite as the conductive skeleton, and graphite and carbon fiber as the conductive additives by a more conventional molded thermosetting two-step method, in which it was found that natural scaled graphite had the highest conductivity due to its high aspect ratio, followed by natural microcrystalline graphite, and artificial graphite had the lowest conductivity. The addition of conductive carbon black effectively improved the electrical conductivity of the bipolar plates, which was 105 S/cm at 180 °C. The addition of 4% carbon fiber increased the flexural strength of the bipolar plates by 29% up to 37 MPa. Therefore, flake NG can be considered as the main conductive filler for composite bipolar plates.

Mara et al [45] prepared a composite bipolar plate prepared from a mixture of phenolic resin, graphite, carbon black and carbon fiber. In this study, the large number of defects and dangling bonds in carbon black and therefore highly susceptible to oxidation increased the corrosion current density and reduced the corrosion resistance of the electrode plates, while carbon nanotubes could compensate for the reduced corrosion resistance of carbon black due to the agglomeration effect, thus greatly improving the electrical properties of the plates. The best performance is achieved at 2 wt.% multi-walled carbon nanotubes, with a conductivity of about 40 S/cm through the plane and a total conductivity close to 200 S/cm, and good corrosion resistance.

S.R. Dhakate et al [46] synthesized expanded graphite based composite bipolar plates by chemical insertion of natural graphite and rapid expansion at high temperature using expanded graphite as raw material. The expansion rate of expanded graphite ranged from 75-100 cc/gm. 50 wt% of composite bipolar plates had a bulk density of 1.50 g/cm3, conductivity >120 S/cm, flexural strength of 54 MPa, Young's modulus of 6 GPa, and Shore hardness of 50. EG/phenolic resin composite BP was prepared by resin vacuum impregnation and hot pressing method. 25% water-soluble phenolic resin impregnated BP has 33.2 MPa tensile strength, 64.9 MPa flexural strength, 8.9 mΩ-cm^2 resistivity, 3×10^{-7} cm^3 cm s^{-2-1} permeability and 4.0 μA/cm^2 corrosion current density, with significantly higher performance than graphite bipolar plates.

Similar to epoxy resin-based composite bipolar plates, carbon fiber is considered to be the best material to improve the performance of phenolic resin bipolar plates.Bo Lv et al [47] prepared phenolic resin-based bipolar plates using carbon fiber treated with Fenton reagent for 2 h. The optimal mass content of carbon fiber was 4%. At this time the maximum power density could reach 662.75 mW cm^{-2} . The flexural strength reached 35 MPa and the electrical conductivity was close to 240 S/cm.

In addition to adjusting and optimizing the ratios of different conductive fillers to obtain inexpensive materials with superior performance, modification of phenolic resins is also an important means to improve the performance and accelerate the processing efficiency of this type of bipolar plates [48]. Ouyang Tao [49] prepared expanded graphite/PF-PVB composite bipolar plates for proton exchange membrane fuel cells by modifying phenolic resin with polyvinyl butyral (PVB) and using expanded graphite as the first conductive filler by molding method. The specimens at a PVB:PF ratio of 1:2 when the mass fraction of modified resin was fixed at 30 wt.% exhibited the best planar conductivity and surface specific resistance of 192.3 S/cm and 47.3 mΩ-cm, respectively^2 . Minkook Kim et al [50] investigated the acceleration of the curing reaction of carbon fabric/phenolic materials with p-toluenesulfonic acid catalyst (strong organic acid catalyst), and on the Based on this, an ultra-high speed curing bipolar plate was developed. The fastest curing speed was achieved at 120°C, and the curing time was reduced from 30 min to 5 min by using 10% catalyst, and the temperature could be set at 100°C considering the deterioration of the properties due to the rapid evaporation of water from the resin at high temperatures. The ASR of the resulting bipolar plates was reduced to 27 mΩ-cm by adding carbon black particles and surface flame treatment^2 , and the tensile strength reached up to 565 MPa. In a subsequent study, the authors developed a continuous process for mass production of this type of modified phenolic resin composite bipolar plates using hot rolling [51], further optimizing the process parameters, and the permeability of the resulting bipolar plates was maintained at 0.

The main surface treatment method for phenolic resin is flame carbonization. minkook Kim et al [52] flame treated the surface of carbon fabric/phenolic composite bipolar plate to carbonize the surface in order to reduce the variation of interfacial contact resistance with treatment time and temperature. As a result, the ASR of the carbon fabric/phenolic composite bipolar plate treated with propane flame for 5 s was reduced by 30% due to the carbonization of phenolic resin. the ASR was 27 mΩ-cm^2 , which was lower than the DOE target value of 30 mΩ-cm^2 . the bending strength also decreased with the increase of flame treatment time. In contrast, the bending strength of the carbon fiber/phenolic composite treated with propane flame for 5 s was 48.6 MPa, which met the DOE target value of 40 MPa. The gas permeability met the DOE target after 5 s of propane flame treatment.

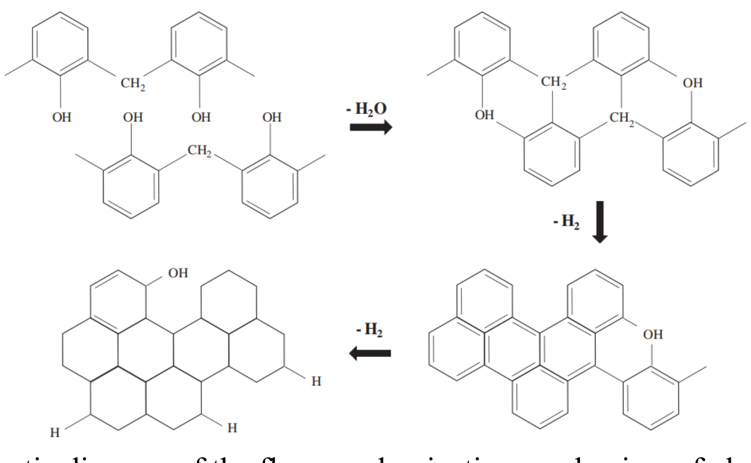

Figure 5.Schematic diagram of the flame carbonization mechanism of phenolic resin [52]

2.3 Thermoplastic material-based bipolar plates

Thermoset composites have been studied earlier and their applications are relatively mature. However, the disadvantage of thermoset composites is that they cannot be recycled, which causes serious environmental and cost-efficiency problems. Meanwhile, compared with thermoset composites, thermoplastic composites have the advantages of high fracture toughness, high impact resistance, recyclability, high productivity (in situ consolidation), infinitely long storage period, and easy welding and repair, making them one of the first choices for future large-scale preparation of bipolar plate composites [53].

Tao Yang et al [54] prepared thermoplastic composite bipolar plates based on polyphenylene sulfide (PPS) and seconal microbeads (MCMB) at 40 MPa and 400 °C. The bipolar plates performed best when the molding time was 30 min and the content of PPS was 20%, with an in-plane conductivity of 133.7 cm/S, a through-plane conductivity of 21.37 cm/S, and a flexural strength of 38.82 MPa.

Thermoplastic composites for bipolar plate applications usually consist of two steps: preparation of thermoplastic prepreg and preparation of thermoplastic laminate. Therefore, there are two main reasons for the poor interlaminar mechanical properties of the material: first, the weak interfacial bonding between the thermoplastic prepreg fibers and the plastic; and second, the absence of fiber-reinforced non-conductive rich aggregation zones between the thermoplastic laminate layers. Many methods have been devised by related scholars to solve the above two problems in order to investigate more mature thermoset composites. In the first problem, it can be solved by changing the fiber surface, such as improving the molding conditions, heat treatment, etc. For the second problem, the and delamination resistance of the laminate is improved by enhancing the resin matrix, such as doping with metals, covalently bonded compounds, carbon nanotubes, and carbon fibers, while carbon nanotubes can also improve the electrical conductivity of the polar plates [55].

The addition of covalent bonding compounds can effectively improve the bending strength of thermoplastic bipolar plates.Mei Bingchu et al [56] prepared a new composite bipolar plate using polyvinylidene fluoride as the binder and titanium carbide as the conductive filler by compression molding process. The conductivity and bending strength of the composite bipolar plate were 28.83 cm/S and 24.92 MPa, respectively.At this time, the Ti SiC_{32} content was 80% wt. and the prepared mold pressure was 10 MPa with 10 min compression.The performance can be further improved by optimizing the Ti_3 SiC_2 content, Ti_3 SiC_2 particle size and compression molding conditions, especially the mold pressure.Kwang Sang Park et al [57] prepared composite bipolar plates based on fluorinated ethylene propylene and graphite nanoplatelets (GnP) or graphite flakes (GF) decorated with fine silicon carbide particles as fillers using compression molding method. Due to the homogeneous dispersion of the silicon carbide particles and the strong chemical bond between GF (or GnP) and silicon carbide particles, the electrode plates have a high mechanical strength, the highest being 55 MPa for FEP/GnP/SiC, but the addition of SiC leads to a decrease in conductivity. The conductivity can reach up to about 700S/cm.

The addition of carbon materials or metals with high aspect ratios is also a means to enhance the performance of thermoplastic bipolar plates.Bin Hu et al [58] prepared high-performance polyvinylidene fluoride/graphite/multiwall carbon nanotube composite BPs with separated conductive networks by structural design and compression molding, which greatly improved their low conductivity due to the formation of "brick-mud" structure in the composite bp by structural manipulation. The conductivity of the composite BP with low filler content was greatly improved due to the formation of a "brick-mud" structure in the composite bp by structural manipulation. In addition, separated synergistic conductive networks were observed in the composite BP after the addition of MWCNTs. The electrical conductivity at a dosing ratio of 5 wt% MWCNTs and 35 wt% graphite was 161.57 S/cm, with an area specific resistance of 7.5 mΩ-cm^2 , a bending strength of 42.65 MPa, and superior hydrophobic and corrosion resistance properties.

Sirawit Witpathomwong et al [59] investigated the effect of adding 0-2 wt% of carbon nanotubes to a matrix of graphene and benzoxazine with constant content of graphite on the properties of the resulting composites. The carbon nanotubes have a very high aspect ratio, resulting in improved thermal conductivity in all three directions. In addition, carbon nanotubes can contact with adjacent graphene and graphite through bridging, forming an effective conductive network and improving the electrical conductivity. The composite with 2% of carbon nanotubes has a high transverse thermal conductivity of 21.3 W/mK, which is 44 times higher than that of carbon nanotubes. In addition, the composite has a conductivity of 364 S/cm, a bending strength of 41.5 MPa, and a modulus of 49.7 GPa.

Nabilah.A.M. Radzuan et al [60] premixed ground carbon fiber reinforced polypropylene with carbon nanotubes or graphene nanosheets through an extrusion process to orient the fibers, and then molded at 13.8 MPa and 200°C for 15 min. when the MCF content was 70 wt%, the through-plane conductivity of the composites was 14.8 S/cm, which was higher than 4.9 S/cm of the xGnP composite. the bending strengths of the CNT and x GNP compressed at 70 wt% were increased to 99.6 MPa and 172.5 MPa, respectively. the authors improved the structure and composition of the pole plates in a subsequent study to be suitable for high temperature environments at 400°C [61].

Aninorbaniyah Bairan et al [62] developed PEMFC bipolar plates by compression molding using polypropylene as polymer matrix and graphite, carbon black and carbon nanotubes as reinforcement materials. The addition of carbon nanotubes as a third filler in the G/CB/CNT/PP composites resulted in a synergistic effect to improve the electrical conductivity, bending strength, bulk density and hardness of the composites. This is because the synthetic graphite particles contain a large number of flaky interstices between them, which can be efficiently filled by carbon nanotubes. The electrical conductivity was up to 158.32 cm/S and the bending strength was 29.86 MPa.

2.4 Inorganic salt cement matrix bipolar plate

Phosphate and aluminate cements are extremely stable in low pH environments, and their dense structure provides an excellent barrier to gas penetration. Compared to organic materials, inorganic cements have better corrosion resistance and lower cost.

Wenbin Hao et al [63] developed a magnesium phosphate cement (MPC) composite bipolar plate applied to direct methanol PEMFC using a simple hot pressing process to solve the above problem. The material is an MPC composite composed of MPC, which partially replaces fly ash (FA) as the bonded matrix phase, and multifaceted carbon-based materials (including graphite, carbon fibers, and multi-walled carbon nanotubes) as the conducting phase. The optimized electrode plates had conductivity close to 140 S/cm and flexural strength close to 23 MPa.The researchers prepared modified bipolar plates of magnesium phosphate cement composites using a hot-pressing assisted hydration process in 2019 [64]. It was shown by tests that the fuel cell stack consisting of three single cells connected in series achieved a fairly good maximum current density of 79.78 mA/cm^2 , and peak power density of 25.54 mW/cm^2 at 80 °C. In addition, good cyclability performance was observed by switching tests, and the fuel utilization test was operated for 3.5 h at a discharge load of 25 mA/cm .^{2}

The biggest drawback of this type of bipolar plate is the weak bending strength, and the bending strengths of the above bipolar plates are all difficult to reach the target values specified by DOE. This is due to the microstructure and processing characteristics of inorganic salt cements, so this type of bipolar plate is mostly used in direct methanol PEMFC, which requires less bending strength.

2.5 Other studies of composite bipolar plates in single-phase matrix systems

In addition to the above matrix systems, some studies have developed composite bipolar plates with other materials such as cyanate, phenolic epoxy resin, milamine foam (MF), nylon, and PEEK resin as the matrix.

Carbon/epoxy composite bipolar plates have high specific strength and specific stiffness, which are ideal replacements for brittle graphite bipolar plates for lightweight proton exchange membrane fuel cells (PEMFC). However, conventional carbon/epoxy composite bipolar plates are not suitable for high-temperature proton exchange membrane fuel cells (HT-PEMFC) because the operating temperature of high-temperature cells is higher than the glass transition temperature of conventional epoxy resins, so researchers have investigated other composite bipolar plates suitable for high-temperature operating environments.Dongyoung Lee et al [65,66] developed a composite bipolar plate using randomly oriented nonwoven carbon felt and cyanate modified epoxy resin. In this study, the cathode and anode were integrated into a single bipolar plate to reduce the thickness and weight of the HT-PEMFC power stack. The developed composite bipolar plate was treated by the "soft-layer method" and exhibited excellent electrical properties due to the exposure of bare carbon fibers on the surface, and no additional surface treatment or coating was required to improve production efficiency. At a compaction pressure of 1.38 MPa, the ASR of the composite bipolar plate is only mΩ-cm^2, which greatly meets the U.S. Department of Energy (DOE) target of 20 mΩ-cm^2. The creep test results show that the composite bipolar plates are dimensionally stable under HT-PEMFC operating conditions. The thickness and mass of the single cell are only 1.7 mm and 1.7 g, respectively, with a power of 11.2 W and a power density of 0.45 mW /cm^2 in the operating temperature range of 140-160°C.

The presence of phenolic hydroxyl and hydroxymethyl groups in phenolic resins can lead to brittle resins, and the structure of phenolic resins was adjusted by epoxidation, and the resulting phenolic epoxy resins combine some of the advantages of epoxy and phenolic resins with higher toughness and heat resistance.Chen Hui et al [67] chose a low-cost phenolic epoxy resin mixed with natural graphite and black carbon, and prepared graphite/ polymer composite bipolar plates. The best performance composite bipolar plates were obtained when the processing conditions were: resin content of about 15 wt.%; molding pressure of 200 MPa; curing temperature of 180°C; graphite particle size of 200 mesh, the electrical conductivity could reach 120 S/cm and the bending strength was higher than 38 MPa.

Xiaoyu Mao et al [68] developed a composite bipolar plate with EG/Ni@ honey amine foam/EG sandwich structure using a lamination method to prepare it. In this study, the extremely porous MF structure provided enough space for epoxy resin immersion, and nickel was absorbed by the porous MF sponge to form a complete conductive path, which also improved the electrical conductivity to 320 S/cm and a bending strength of 56 MPa, both of which were much higher than that of typical EG bipolar plates. The amount of resin enriched in the interlayer also ensures a low gas permeability (2.16 × 10-9Std-cm^3 /s-cm^2 -Pa). In addition, the residual resin on the surface of the polar plates is smaller in size and therefore its wettability is better.

Wenkai Li et al [69] prepared PEEK resin bipolar plates by uniformly dispersing multi-walled carbon nanotubes on the surface and pores of expanded graphite by in situ vapor deposition. The agglomeration problem was effectively avoided, and the properties of the composite BPs were effectively improved by the synergistic effect with graphite. The modified composite bipolar plates with the best electrical conductivity of 334.53 S/cm and flexural strength of 50.24 MPa at 2% addition of in situ deposited carbon nanotubes can meet the 2025 DOE requirements in all properties.

Jenn-Kun Kuo et al [70] used a composite material consisting of nylon-6 and S316L stainless steel alloy fibers to fabricate a bipolar plate by an injection molding process. The electrode plate is inexpensive and has good stability. However, the electrical properties of this bipolar plate were poor, with a conductivity of only 60 S/cm and a current density of 350 mA/cm^2 . Therefore, further research is needed to improve the electrical properties.

3. Multi-phase matrix system composite bipolar plate

In contrast to single-phase matrix composites, it has been found that multi-phase polymer blends can reduce the particle loading because the conductive filler can be selectively located at the interface between multiple polymer phases or in one of the polymer phases. When the distributed conductive fillers reach the percolation threshold, a continuous structure of conductive fillers is formed in the polymer, called the multiple percolation effect. This multiple percolation effect can greatly reduce the percolation threshold of the material and enable the formation of a continuous conductive pathway in composites with only a small amount of conductive filler, which greatly improves the electrical conductivity while reducing the raw material cost [71,72]. Using the double osmosis method as the underlying theory, researchers have developed multiphase matrix composite bipolar plates with extremely high electrical properties.

3.1 Double osmosis method

Sumita et al [73] first investigated the electrical conductivity of mutually incompatible carbon black-filled polymer blends in 1992 in relation to the polymer blending ratio and found the double osmosis effect.Tchoudakov Rosa et al [74] prepared composites of CB particles and polypropylene/nylon (PP/Ny) immiscible blends by molding and investigated their resistivity and morphology. The study was carried out. The double osmosis effect was observed in the experiments, and the principle of the double osmosis effect was concluded based on the experiments. Firstly, the different affinity of CB for the blends leads to a preferential distribution of CB in one of the phases, and a very fast transfer rate of CB to the preferred phase at high temperatures is required.The double osmosis effect due to the selective distribution of short carbon fibers was first discovered by C. Zhang [75].Man Wu et al [76] studied and analyzed the microstructure, mechanical properties and electrical conductivity of four polymer blends filled with carbon nanotubes, mechanical properties and electrical conductivity of four polymer blends filled with carbon nanotubes. It was concluded that the distribution of carbon nanotubes in the polymer blends was mainly determined by the wetting coefficient and processing conditions, and the double-percolation effect occurred when most of the carbon nanotubes were distributed in one polymer phase while the other polymer phase remained neat. And the higher the flexural strength of the polymer in the neat phase, the higher the overall flexural strength of the composite. However, cracks and bubbles that may form during the injection molding process can reduce the strength of the pole plate.Hiroshi Yui et al [77] investigated the effect of carbon black (CB) addition on the microstructure of injection molded high density polyethylene (HDPE)/polypropylene blends. It was demonstrated that the composite blend material can self-assemble to form a random conductive network even in a very short period of time and at high shear rates. Researchers have developed a variety of new bipolar plates with much higher performance than the composite bipolar plates of single-phase matrix systems based on the theoretical basis of the double-permeability effect [83-88].

Translated with www.DeepL.com/Translator (free version)Man Wu et al [78] fabricated a composite bipolar plate with a tri-continuous structure by injection molding at low cost using carbon nanotubes filled with polyethylene terephthalate (PET)/polyvinylidene fluoride blends. The PNT-filled PET/PVDF composite bipolar plate showed a 2500% increase in electrical conductivity, 36% increase in tensile strength, and 320% increase in elongation at the same carbon loading.

Ha Eun Lee et al [79] developed PEMFC bipolar plates by sol-gel impregnation method using compression molded carbon fiber/polypropylene/polyethylene composites. This study improved the degree of impregnation and dispersion of multi-walled carbon carbon nanotubes without increasing the viscosity. The prepared bipolar plates had a maximum conductivity greater than 120 S/cm, an ASR around 27 mΩ-cm^2 and a bending strength greater than 41 MPa, which met the objectives set by DOE. The authors prepared the carbon nanotube and carbon felt-reinforced PP/PE composite plate by sol-gel impregnation and double percolation methods in the subsequent study, which further enhanced the bipolar plate performance and made it have better thermal stability. The ASRs of this carbon fiber-reinforced composite BP and carbon felt-reinforced composite BP were 25.3 mΩ-cm^2 and 19.4 mΩ-cm^2 , respectively [80].

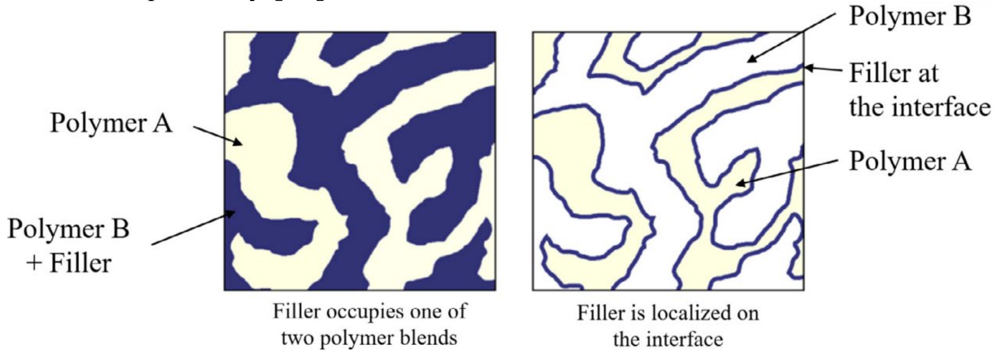

Fig. 6. packing distribution in double osmosis theory [36]

Fangfang Zou et al [82] established a well-hardened conductivity network of double-permeable PCL/PS/MWCNT composites under CO2-assisted annealing conditions, which improved the conductivity and EMI shielding performance. After supercritical CO2 annealing, the percolation threshold decreased from 0.50 wt% to 0.24 wt% and the EMI SE increased from 31.8 dB to 39.8 dB, an improvement of 25%. The conductivity is close to 700 S/cm, much higher than other materials suitable for composite pole plates.

In summary, the composites prepared by using the phenomenon of selective distribution of conductive fillers in uncoordinated polymers have very high electrical conductivity and good mechanical strength and heat resistance, electromagnetic shielding and other properties. However, there are still quite few studies discussing how to produce this composite commercially, on a large scale and at low cost. Meanwhile, there is still much room for improvement in processing methods. Therefore, the preparation of composite bipolar plates by the double osmosis method is considered to be the key research direction for future composite bipolar plates.

4. Summary

Composite bipolar plates have become a popular research direction in the field of bipolar plates in recent years due to their better mechanical strength than graphite bipolar plates, better corrosion resistance than metal bipolar plates, and lower cost, flexible raw material ratios, and applicability to a variety of different working environments. The main matrix materials used in composite bipolar plates are epoxy resin, phenolic resin, thermoplastic and other materials such as cyanate and inorganic salt cement. Current research focuses on improving the electrical and mechanical properties of composite bipolar plates, while some other research focuses on reducing the production cost of plates, shortening the processing time, improving the corrosion resistance of plates, and improving the structure of plates.

Typical epoxy resin-based composite bipolar plates use graphite and carbon black as conductive fillers. Recent studies have found that doping with carbon nanotubes, carbon fibers and other materials with high aspect ratios can reduce the over-permeability threshold of the material, thus greatly improving the electrical and mechanical properties of the plates. Some studies have also concluded that the doping of metal foil can obtain a better structure and thus improve the overall performance of the plates. The improvement means of epoxy resin bipolar plate include improving the bipolar plate structure, surface modification, coating, and improving the processing process. The soft-layer method is considered to be a better improvement method to improve the electrical properties of the electrode plate.

Phenolic resins have the advantages of better heat resistance, shorter curing time, and lower raw material prices than epoxy resins, so phenolic resins can be used as a matrix material for high-temperature bipolar plates and are more suitable for rapid mass production. However, phenolic resins require heating and pressure during curing; there is water as a by-product of the reaction, so there are bubble residues and the disadvantage of large curing volume shrinkage; their flexural strength is lower than epoxy resins, so recent literature focuses on improving the flexural strength and air tightness of phenolic resin bipolar plates. The main means of improvement are adjustment of filler type and ratio, improvement of processing, surface treatment, chemical modification of phenolic resin, etc.

Thermoplastic materials have the advantages of high fracture toughness, high impact resistance, recyclability, high production efficiency (in-situ consolidation), infinitely long storage period, easy welding and repair, etc., which are suitable for application in the production of composite bipolar plates. However, the interlayer mechanical strength of thermoplastic pole plates is poor due to the weak interfacial bonding between the thermoplastic prepreg fibers and the plastic; secondly, there is no fiber-reinforced non-conductive rich aggregation zone between the thermoplastic laminate layers. Solutions to the first problem include improved molding conditions, heat treatment, etc. The second problem can be improved by enhancing the resin matrix, such as doping with metals, covalent bonding compounds, carbon nanotubes, carbon fibers, etc., which can improve the laminate's and anti-delamination properties, while conductive materials such as carbon nanotubes can also improve the electrical conductivity of the pole plate at the same time.

Other materials that can be used as composite bipolar plate substrates include inorganic salt cement, cyanate, dense amine foam, nylon, etc. Phosphate and silicate cements are able to have high stability in low pH environment, while their dense structure provides excellent barrier to gas permeation; compared with organic materials, inorganic cements have better corrosion resistance and low cost. However, its bending strength is poor, so it is often used in direct methanol proton exchange membrane cells. Since the working temperature of high-temperature cells is higher than the glass transition temperature of traditional epoxy resin, cyanate is considered a reliable material for the preparation of high-temperature composite bipolar plates; by adjusting the structure of phenolic resin through epoxidation, the resulting phenolic epoxy resin has some advantages of both epoxy resin and phenolic resin, with higher toughness and heat resistance, and is also one of the optional matrix materials for composite bipolar plates. In addition, other materials suitable for composite bipolar plates include dense amine foam and nylon, etc. The composite plates prepared with these materials can meet the target performance set by DOE.

This paper also introduces a double-permeation method for improving the properties of composites. In multiphase polymer blends, conductive fillers can be selectively located at the interface between multiple polymer phases or in one of the polymer phases. When the distributed conductive fillers reach the superpermeability threshold, a continuous structure of conductive fillers is formed in the polymer, which is known as the multiple superpermeability effect. This multiple percolation effect can greatly reduce the material's percolation threshold, enabling the formation of a continuous conductive pathway in the composite with only a small amount of conductive filler, greatly increasing the conductivity while reducing the cost of raw materials. The composites prepared by using the selective distribution of conductive fillers in unblended polymers have very high conductivity and good mechanical strength and heat resistance, electromagnetic shielding and other properties. However, there are still quite few studies discussing how to produce such composites commercially, on a large scale and at low cost. Meanwhile, there is still much room for improvement in processing methods. Therefore, the preparation of composite bipolar plates by the double osmosis method is considered to be the key development direction of composite bipolar plates in the future.

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