Progress of bipolar plates for proton exchange membrane fuel cells

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Abstract. The basic structure of proton exchange membrane fuel cell mainly consists of bipolar plates, diffusion layer, catalyst layer and proton exchange membrane, of which the bipolar plates are the most important components. In recent years, composite bipolar plates have become the focus of bipolar plate material research because they combine the advantages of graphite bipolar plates and metal bipolar plates. Therefore, this review explains the development of composite bipolar plates in recent years in a clearer way by sorting out and analysing the pairing of different carbon and resin materials, which will be helpful for bipolar plate development.

Keywords: Bipolar Plates; Composite Bipolar Plate; Carbon material; Resin material; Matching..

1. Introduction

The basic structure of proton exchange membrane fuel cell is mainly composed of bipolar plate, diffusion layer, catalyst layer and proton exchange membrane as shown in Fig. 1[1], of which the bipolar plate is the key component with the most important function, and its structural weight accounts for about 80% of the total amount of the device, and the cost accounts for about 40% of the whole device. The proton exchange membrane fuel cell uses solid electrolyte membrane as electrolyte, the fuel can be pure H2 or methanol, natural gas and other reformed H2-rich mixture, the oxidant is generally pure O2 or air. After the fuel enters the battery, it diffuses to the interface between the anode catalyst layer and the membrane, and loses electrons in the oxidation reaction under the action of the catalyst, and the H+ formed is conducted through the membrane to the interface between the membrane and the cathode, and reacts with the oxidant under the action of the cathode catalyst, and the water generated is discharged from the cathode chamber in the form of water vapour or condensate, and the electrons generated by the anodic reaction reaches the cathode after the external circuit has done the work on the load.

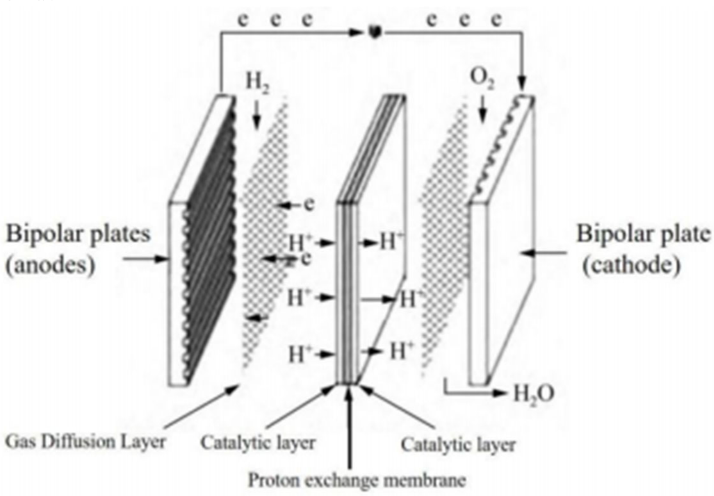

Fig. 1 Structure of proton exchange membrane fuel cell (PEMFC)[1]

Bipolar plate is one of the most important components of fuel cell, which distributes oxidant and reductant, achieves good current conduction between single cells, brings out the residual heat in the reaction zone, and prevents the leakage of gas and coolant in the fuel cell stack.

Carbon and resin materials have a wide range of applications in the preparation of composite bipolar plates. Carbon material becomes an important part of composite bipolar plates because of its high conductivity, high strength and excellent corrosion resistance. The resin material, on the other hand, mainly assumes the role of binder to improve the mechanical properties and durability of the composite material. By reasonably designing the structure of composite bipolar plates, their performance can be further improved and efficient and low-cost preparation can be achieved.

In recent years, because the composite bipolar plate combines the advantages of graphite bipolar plate and metal bipolar plate, and at the same time has good electrical conductivity, mechanical strength, corrosion resistance, as well as good processing performance and low cost, it has become the focus of research on the bipolar plate materials for proton exchange membrane fuel cells[2], therefore, this review explains the development status of the composite bipolar plate in recent years more clearly by collating and analysing the different matching of carbon and resin materials, and provides a clearer explanation for the future development of proton exchange membrane fuel cells in the future. Therefore, this review provides a clearer description of the development status of composite bipolar plates in recent years by collating and analysing different carbon and resin materials, which will provide some guidance for the development of proton exchange membrane fuel cell bipolar plates in future.

2. Carbon material

Influence of Graphite Types on the Performance of Composite Bipolar Plates Numerous studies have shown that the performance of the graphite powder used in the preparation of fuel cell bipolar plates is a key factor in ensuring that the bipolar plates achieve the desired performance. Based on this,a classification of different carbon materials is explained and synergies between different carbon materials are collated.

2.1 Study of composite bipolar plates with carbon fibres as conductive fillers

Carbon fibres possess high modulus, high strength, low density and good electrical conductivity, making them ideal fillers for the preparation of composite bipolar plates. Studies have shown that increasing the carbon fibre content can enhance the electrical conductivity and mechanical strength of bipolar plates. However, too high a content reduces processability and increases cost. Therefore, it is important to investigate how to reduce the carbon fibre content and improve the processability while maintaining the electrical conductivity and mechanical strength.

Mohd Yusuf Zakaria[3] will synthesise carbon composite bipolar plates consisting of graphite and milled carbon fibres as conductive filler and epoxy resin as polymer matrix, these carbon composites were developed using compression moulding. For composites containing 2 wt% carbon fibres (CF) with a filler loading of 80 wt.%, the highest electrical conductivity obtained from said materials was 69.8 S/cm, with an in-plane conductivity of 69.8 S/cm, and an in-plane conductivity of 50.34 S/cm.This value is 30% higher than that obtained for atypical graphite/epoxy composite with a filler loading of 80 wt.%, with an in-plane conductivity of 53 S/cm and in-plane conductivity of 40 S/cm. The flexural strength increased to 36.28 MPa compared to a single filler system of approximately 25.22 MPa. The study also found that the Generalised Effective Medium (GEM) model was capable of predicting the in-plane and in-plane conductivities of both the single filler and the multiple filler composites.

Kang[4] designed and fabricated ultrathin composite bipolar plates for fuel cells by taking advantage of the excellent mechanical strength and in-plane conductivity of carbon fibre and epoxy prepregs. They used amultilayer bipolar plate structure in which prepreg, pure graphite and graphite resin composite layers were combined and compression moulded. The multilayer prepreg bipolar plates are about 0.6 mm thick and have good electrical properties (in-plane conductivity of 172 S/cm and in-plane conductivity of 38 S/cm) with high-quality serpentine runner configurations. The experimental results clearly show that their proposed bipolar plate design and fabrication greatly improves the low through-face conductivity and poor formability of the original prepreg.

2.2 Study of composite bipolar plates with expanded graphite (EG) as conductive fillers

Expanded graphite (EG) is considered as a potential filler for the preparation of composite bipolar plates due to its high electrical conductivity, low density and good mechanical strength. It was found that the electrical conductivity and flexural strength of composite bipolar plates increased accordingly with the increase of expanded graphite content. However, too high content of expanded graphite leads to poor processing performance and increased cost of composite bipolar plates.

Piotr Rzeczkowski[5] studied the conductive fillers of expanded graphite melt compounded with polypropylene (PP) with contents between 10-80 wt%. Resistivity, thermal conductivity and mechanical properties varied with filler content. Electrical and thermal conductivity increased with increasing filler, but tensile and flexural strength decreased. Young's modulus and flexural modulus increased with increasing filler content. Thermogravimetric analysis verified the actual filler content. Wettability and adhesion were assessed by contact angle measurements, which indicated that surface tension increased with filler content. Joint strength of graphite composites, assessed in lap shear tests after adhesive joining, was enhanced with increasing filler content. Surface treatments (plasma and chemical) significantly improved surface tension and tensile friction shear strength. Graphite prepreg hybrid inner layers were prepared and the electrical conductivity was improved by composite outer layers. The in-plane and trans-plane conductivities of the composite bipolar plates were 172 S/cm and 38 S/cm, respectively.

For the newnanofiller modification, 2023 Li[6] in synthesized a continuous reticulated carboxylated multi-walled carbon nanotube (MWCNT) coating on the surface of expanded graphite using chemical vapor deposition (CVD) and carboxylation modification, and prepared composite bipolar plates by moulding using reticulated carboxylated MWCNT, expanded graphite and resin. By optimising the carboxylation treatment time and nanofiller content, the composite bipolar plates had the best performance at a carboxylation treatment time of 15 min and a filler content of 2.4%. The planar conductivity reached 243.52 S/cm and the flexural strength was increased to 61.9 MPa. The thermal conductivity and hydrophobicity were improved compared to the conventional graphite/resin composite bipolar plates, and they showed good corrosion resistance under the PEMFC operating environment. Their work provides a new paradigm for nanofiller modification of bipolar plates.

The above section focuses on the application of conductive fillers such as carbon fibre and expanded graphite in the study of composite bipolar plates. Carbon fibre and expanded graphite have received much attention in composite bipolar plate research due to their excellent mechanical strength and electrical conductivity. With the continuous development of new nanofillers, the performance of composite bipolar plates is expected to be further improved in the future.

2.3 Synergies between different carbon materials

Synergy between different carbon materials means that they work together and promote each other to obtain better properties and applications. These carbon materials include graphene, carbon nanotubes, carbon fibres, etc., each with unique properties and applications. Synergistic interactions can enhance the mechanical properties of the materials, such as strength and hardness, as well as their electrical conductivity, allowing for a wider range of applications, such as in electronic devices. This synergistic effect can make use of the strengths of each carbon material to compensate for the shortcomings and achieve a comprehensive improvement in performance.

2022 Li[7] prepared a uniformly dispersed multi-walled carbon nanotube network by in-situ vapour deposition on the surface and pores of expanded graphite, which effectively avoided the agglomeration problem through synergistic interaction with graphite and effectively improved various properties of composite bipolar plates. With the incorporation of 2% in-situ deposited carbon nanotubes, the modified composite bipolar plates have the best conductivity (334.53 S/cm) and flexural strength (50.24 MPa), and all the properties can meet the DOE requirements for 2025.

Modification of composite bipolar plates using in-situ deposition of carbon nanotubes is a feasible route because it can generate multi-walled carbon nanotubes in large quantities, avoiding the agglomeration phenomenon caused by the addition of nanofillers. It can also significantly improve the performance of composite bipolar plates and achieve high performance of composite bipolar plates at a much lower cost.

2023 Muhammad Tariq[8] in this experimental study, conductive polypropylene (PP) composites were prepared by adding conductive fillers such as carbon nanotubes (MWCNT), expanded graphite (EG), and carbon black (CB) by using melt composite technique in twin-screw extruder. Conductive fillers were incorporated in binary, ternary and quaternary formulations to synergistically improve the electrical conductivity and flexural strength of the composites. The effect of filler content at three levels of variability was investigated using a full factorial design. The optimum response parameters of the composites at MWCNT content of 4 wt.%, CB content of 5 wt.% and EG content of 30 wt.% were 39.6 S/cm for conductivity through the surface and 29.4 MPa for flexural strength.

In 2023, Huang[9] prepared multilayer high-performance polybenzoxazine/expanded graphite composite bipolar plates with a "graphite-composite-graphite" structure by compression moulding. Graphite paper and polybenzoxazine/expanded graphite composites were hot-pressed to form a surface graphite layer and a conductive network. The results show that the multilayer synergistic effect has led to an in-plane conductivity of 278.85 S/cm-1, an area specific resistance of 9.70 mU/cm2, and a flexural strength of 75.75 MPa, and that the graphite-composite-graphite structure can increase the power density of a single cell assembled with multilayer composite bipolar plates by 111.02% and 111.02%, respectively. The "graphite-composite-graphite" structure can increase the power density of single battery assembled with multilayer composite bipolar plates by 111.02% and 113.80%, respectively.

Carbon materials play a key role in a number of fields, and when combined with other substances, they may have synergistic effects, resulting in improved performance or new functional properties. Such synergistic effects show great potential and application prospects in many areas. With further research and technological development, more innovations and applications based on the synergistic effects of carbon materials are expected in the future.

3. Resin material

Polymer resins are mainly classified into two categories: thermoset and thermoplastic[10] . Thermosetting resins are more commonly used in the preparation of composite bipolar plates with high hardness, good stability and low viscosity. Common thermosetting resins are epoxy resin, vinyl ester resin and phenolic resin. Thermoplastic resins, whose service temperature is related to the glass transition temperature, commonly include polypropylene, polyethylene, and nylon resins. Both can be used as polymer materials for bipolar plates, but thermosets have long moulding cycles and are not as costly or productive as thermoplastics. Although thermoplastic composites have many advantages, the low electrical conductivity and the need to add conductive fillers to improve electrical conductivity may lead to a decrease in mechanical properties. Therefore, how to balance the electrical and mechanical properties is a challenge in the preparation of bipolar plates.

3.1 Study of composite bipolar plates using phenolic resin as binder

Phenolic resin is ideal for the preparation of composite bipolar plates due to its heat and corrosion resistance and high mechanical properties. When prepared, it can be mixed with conductive fillers (e.g. carbon fibres, carbon nanotubes, graphite, etc.) to form a conductive network and increase the electrical conductivity. At the same time, phenolic resin can also enhance the mechanical properties of composites, such as flexural and tensile strength.

R.K.Gautam[11] used microwave irradiation to prepare exfoliated graphite with a maximum exfoliation volume of 570±10 mL in a few minutes. Phenolic resin composites containing 10-35 wt.% exfoliated graphite, 5 wt.% carbon black and 3 wt.% graphite powder were prepared by compression moulding. The conductivity, bulk density, compressive strength, flexural strength, energy storage modulus, microhardness, and water absorption of the composites were 374.42 and 97.32 S/cm, 1.58

g/cm, 70.43 MPa, 61.82 MPa, 10.25 GPa, 73.23 HV, and 0.22%,respectively.The I-V characterisation showed that the exfoliated graphite/carbon black/graphite powder/resin composite double electrode plates have better performance in cellular fuel cells. This composite plate has low packing density, high electrical conductivity, and good thermal stability, which meets the U.S. Department of Energy's goal to be used as a bipolar plate in proton exchange membrane fuel cells.

Yean-Der Kuan[12] presented agraphite woven fibre composite bipolar plate made by premixing phenolic resin with graphite powder. Due to the low conductivity of the resin region between the fibres, a carbonisation process was used to remove part of the resin to enhance the electrical conductivity. The carbonised bipolar plates reached the DOE 2020 target for flexural strength. Experiments show that the carbonisation process improves battery performance, and increasing the number of carbonisations is even better. Connecting thin copper plates also improves performance. Compared with graphite bipolar plates, the carbonised graphite composite bipolar plates have higher PEMFC performance and have passed a 12-hour stability test, showing good application prospects.

3.2 Study of composite bipolar plates with epoxy resin as binder

Epoxy resin is a commonly used binder with high strength, wear resistance and electrical insulation properties. When compounded with conductive fillers, high performance composite bipolar plates can be formed. The properties of the composite can be optimised by adjusting parameters such as the type of epoxy resin, ratio and curing conditions.

In 2022, Hu[13] prepared epoxy resin-based composite bipolar plates characterised by a 3D graphite structure (3D graphite). A 3D graphite skeleton was formed by mixing, pressing and heating graphite and NH4HCO3 at 90℃ . Subsequently, this skeleton was impregnated with epoxy resin to obtain the 3D graphite/epoxy composite. The composite bipolar plate has excellent electrical conductivity, with in-plane conductivity up to 212.64 S/cm and thermal conductivity up to 16.01 W/mK, which is superior to randomly distributed graphite/epoxy composites. Compared with the conventional hybrid graphite/epoxy composite bipolar plate, the 3D graphite/epoxy composite bipolar plate PEMFC performance was improved by 317.52%, with a maximum power density of 853.42 mW/cm. in addition, the material has good corrosion resistance and hydrophobicity. This study provides a new way for the preparation of PEMFC high-performance polymer-based composite bipolar plates.

José Diaza[14] investigated the feasibility of using surface-enhanced flake graphite (SFG) as a primary filler in an epoxy resin aimed at constructing polymer electrolyte fuel cell (PEFC) bipolar plates. The flexural and tensile strengths of the samples were measured with different ratios of SFG and nanographite (NG) as secondary fillers through a hot-pressing fabrication process. The experimental results showed that the bending stress values of the samples ranged from 30 ~ 19 MPa and the average tensile stress was 13 MPa when the SFG content of epoxy resin (EP) was 40% and 50%.In addition, the highest conductivity value was 1.95 S/cm, which was comparable to that of commercial bipolar plates.

3.3 Study of composite bipolar plates using polypropylene as binder

As a commonly used polymer material, polypropylene has good mechanical properties, chemical stability and processing properties. As a binder, polypropylene can effectively combine different materials to form composites with excellent performance. In the application of bipolar plates, polypropylene binder can enhance the strength and durability of bipolar plates, improve their corrosion resistance and electrical conductivity, thus prolonging the service life of fuel cells. The study of composite bipolar plates with polypropylene as binder is of great significance for improving the performance and life of fuel cells.

Roman Bühler[15] has developed innovative polymer-based composites for fuel cell bipolar plates that need to fulfil the requirements of electrical, thermal and mechanical conductivity and be easy to produce industrially. The team added a variety of conductive fillers such as graphite, carbon black,

carbon fibres, carbon nanotubes and expanded graphite to the polypropylene matrix. The samples were tested for electrical and thermal conductivity, flexural properties and corrosion resistance, and the effects of plate thickness and filler composition were investigated. In addition, electrode area and applied pressure were found to have a significant effect on conductivity. The materials showed good processability during compression moulding, reaching a maximum conductivity of 46 S/cm, which exceeded results in the literature. The team believes that by optimising the fabrication process, the conductivity values are expected to exceed the target of 50 S/cm.

Polypropylene (PP) nanocomposites combined with multi-walled carbon nanotubes (MWCNT) and carbon nanofibres (CNF) were prepared by C.A. Ramı´rez-Herrera[16] by melt blending. These composites focus on mechanical properties and corrosion resistance for PEMFC bipolar plate applications.The incorporation of MWCNT increases the microhardness, modulus of elasticity, and tensile and flexural strengths of PP.The combined addition of MWCNT and CNF produces higher strength hybrid nanocomposites and maintains their processability. Measurements showed slower degradation in the PEMFC environment. According to DOE objectives, PP/20MWCNT, PP/21.5MWCNT and PP/15MWCNT/15CNF nanocomposites are good candidates for PEMFC bipolar plates.

In practical applications, the high performance composite bipolar plates can be prepared by selecting suitable resins and synergising with conductive fillers according to the requirements. Meanwhile, the interaction mechanism between the resin and the conductive filler is investigated to optimise the performance of the composites to meet the needs of various fields.

3.4 Dual percolation structures formed by different resins

Multiphase polymer blends can be used to reduce the conductive filler content in thermoplastic composites. Phase separation of the polymer blend and selective distribution of the conductive filler in one of the polymer phases allows the production of conductive materials with lower filler concentrations, which is referred to as the "bi-permeability concept"[17] . The bipermeable structure of the structure can further improve the performance of the composite bipolar plates. In addition, by modulating the composition and structure of different resins, precise modulation of the performance of composite bipolar plates can be achieved.

Lv Bo[18] in 2019 used phenolic resin, polypropylene resin and natural graphite to prepare composite bipolar plates that meet the requirements by adjusting the ratio of phenolic resin and polypropylene and using hot pressing process. The experimental results showed that when the graphite content was 80%, the phenolic resin to polypropylene mass ratio was 2:1, the moulding temperature was 180 ℃, the moulding pressure was 100 MPa, and the moulding time was 5 min, the composite plate had good mechanical and electrical conductivity, as well as excellent airtightness, hydrophilic/hydrophobicity, corrosion resistance, and full-cell performance. Therefore, this composite bipolar plate is considered to be a bipolar plate for proton exchange membrane fuel cells with promising applications.

In 2023, Chen[19] introduced epoxy resin as a binder for composite bipolar plates into phenolic resin, by which the abundant flexible chain segments (-CHOH-CH-O-) in epoxy resin were well embedded into the structure of phenolic resin to enhance flexibility. When epoxy:phenolic resin = 4:6, the bipolar plate achieved the highest ever efficiency in terms of flexural strength, reaching 46.2 MPa with the same conductive filler content, which is 1.57 times higher than that of the pristine phenolic resin-composite bipolar plate (29.58 MPa), and higher than the 2025 DOE target (40 MPa), and the other properties were maintained at a high level of performance.

Resin materials have a wide range of applications in the preparation of composite bipolar plates. Different resin materials have different performance characteristics, and the precise regulation of the performance of composite bipolar plates can be achieved by regulating the type, content and structure of the resin. However, in practical applications, resin materials also have some problems, such as long moulding cycle, high cost and relatively poor electrical conductivity. Therefore, how to further improve the performance of resin-based composite bipolar plates and achieve an excellent balance of electrical, mechanical and corrosion resistance properties remains an important issue for our researchers in the field of composite bipolar plate preparation.

4. Summary and outlook

Fuel cell, as a newtype of power generation, is gradually entering the public's view because of its cleanliness and environmental protection. Fuel cell is a kind of electrochemical device that can effectively control the chemical reaction of fuel and oxidant and directly convert the chemical energy into electric energy, which is a kind of energy converter that converts the chemical energy in fuel into electric energy, and it is regarded as the fourth type of power generation method following the thermal power, hydroelectric power and nuclear power; it can solve the two major problems of energy saving and environmental protection at the sametime, and it is one of the most promising sources of energy in the future[20, 21] .

After decades of research and development, fuel cell technology has been in a position to be used commercially, and the main factor leading to its inability to be used on a large scale is that the cost is too high. As one of the key components in fuel cells, how to reduce cost and improve performance is the main problem facing the rapid development of fuel cells[22] .

At present, fuel cells have a good development prospect from a worldwide perspective, and the development of fuel cells will inevitably promote the development of fuel cell bipolar plates and other related products, which will make their application scope more extensive, not only in the military, aerospace (spacecraft) and other fields, but also in the civil field will be greatly developed, such as motor vehicles, drones, emergency backup power, household fuel cells, communication base station backup power, non-disruptive fuel cells, and so on. In addition, fuel cells will also be greatly developed in civil fields, such as motor vehicles, drones, emergency backup power, household fuel cells, communication base station backup power, uninterruptible power supply, and independent power stations in remote areas. Moreover, the application of fuel cells in electric vehicles is also growing rapidly, and fuel cell electric vehicles will become the largest commercial value of new energy vehicles. With the continuous promotion of policies and technological breakthroughs, the global fuel cell vehicle sales will grow rapidly in the next few years, and it is expected that in 2032, the market sales will exceed 5 million vehicles, and the market sales will exceed 250 billion U.S. dollars[20], which is a very good opportunity for the development of fuel cell bipolar plates. Composite fuel cell bipolar plates are inexpensive and have stable performance, but due to the complexity of the process, in-depth research is still needed, as well as to explore the conditions for commercial production.

Within-depth research and technological development, composite bipolar plates will play a greater role in fuel cells and other fields in the future. The synergy between carbon and resin materials will continue to promote the optimisation of the performance of composite bipolar plates, bringing more innovations and applications in new energy, environmental protection and other fields. At the same time, it is also necessary to pay attention to the research progress of other new materials, such as graphene, carbon nanotubes, etc., in order to make breakthroughs in the preparation of composite bipolar plates.

In the future, with the continuous development of science and technology, the application of carbon materials, resin materials and other novel materials in composite bipolar plates will be further expanded. Researchers will continue to explore new preparation methods, new material systems and new structural designs, with a view to improving the performance of composite bipolar plates while reducing the preparation cost and realising high-performance, low-cost fuel cell bipolar plates. In addition, the study of multifunctional composite bipolar plates will also become a major trend in the future, such as the integration of heating and cooling functions in composite bipolar plates to adapt to the needs of different working environments.

In terms of carbon materials, researchers will conduct in-depth studies on the synergistic effects of carbon materials and other materials, with a view to improving the performance of composite

bipolar plates while achieving new functional properties. For example, the synergy of high electrical conductivity, high mechanical properties and low expansion coefficient can be achieved by modulating the composition and structure of carbon materials with resins, conductive fillers and other materials to meet the performance needs of fuel cells at different operating temperatures.

Finally, with the deepening of research and continuous technological innovation, we believe that more composite bipolar plates with high performance, low cost, environmental protection and other characteristics will appear in the future, laying a solid foundation for the development of China's fuel cell industry. At the sametime, the application of carbon materials, resin materials and other materials in emerging fields will also continue to expand, making greater contributions to the sustainable development of human society.

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