



Progress on graphite-based bipolar plates for use in proton exchange membrane fuel cells

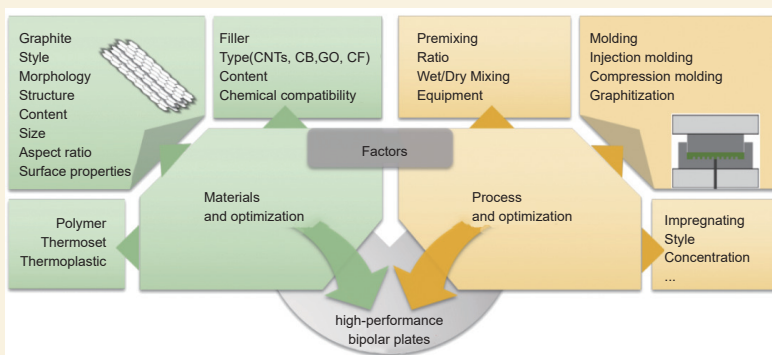
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Abstract: Graphite-based bipolar plates (GBPs) possess excellent electrical and thermal conductivity, corrosion resistance, and durability, making them promising components for proton exchange membrane fuel cells. However, the inherent brittleness and porous structure of graphite can result in inadequate mechanical strength and high gas permeability, thereby limiting the practical application and large-scale manufacture of GBPs. This

review summarizes recent advances in GBP fabrication, focusing on the effects of the graphite raw materials, polymer matrices, conductive and reinforcing fillers, and molding processes on GBP performance. The relationships among graphite particle size, morphology, surface characteristics, filler dispersion, interfacial bonding, resin content, and the electrical conductivity, mechanical strength, gas permeability, and corrosion resistance of GBPs are discussed. The effects of material composition, microstructure, and processing conditions on the overall performance of GBPs are also analyzed. In addition, the trade-offs between electrical conductivity, mechanical strength, gas permeability, corrosion resistance, and processability are highlighted. Finally, strategies for improving performance are summarized, including optimization of the graphite raw materials, multiscale filler design, interfacial regulation, resin-content optimization, and improvements in molding processes.

Key words: Proton exchange membrane fuel cell; Bipolar plate; Filler; Polymer matrix; Process and optimization



1 Introduction

With the increasingly severe environmental problems caused by global greenhouse gas emissions, scientists around the world have turned their attention to the energy transition, from the fossil fuels that emits a lot of CO₂ to the renewable clean energy, building a clean energy application system without greenhouse gas emissions^[1–4]. Proton exchange membrane fuel cell (PEMFC) powered by hydrogen is a potential alternative to replace the traditional energy equipment powered by fossil fuel. In the reaction process, the chemical energy of hydrogen and oxygen can be converted to electricity, only water and heat are discharged^[5–8]. The fuel cell system includes the catalyst^[9–10], bipolar plate^[11–21], proton exchange membrane, gas diffusion layer and other key components^[11–12,22–25], as shown in Fig. 1a. The per-

formance and cost of each part will affect the performance and commercial application of the fuel cell. The bipolar plate is one of the key components of the fuel cell, as the skeleton supporting the fuel cell stack^[20–21,26–30]. It takes up most of the volume, weight (~ 70%) and cost (~ 30%) of the stack, and also plays a role in connecting multiple single cells, bearing the clamping force, dividing the reaction area, transmitting the reaction gases, collecting and conducting electricity (Fig. 1b)^[31]. The high performance and low cost of bipolar plate materials are the key to their application. At present, the market is not perfect and the

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cost of bipolar plate is still high. However, with the commercialization of fuel cells, the cost of each key component will decrease. It is estimated that when the annual production of fuel cell systems reaches 500 000 units, the cost of bipolar plate will account for 28% of that of fuel cell stack, and the cost will decrease further when the market becomes more mature (Fig. 1c)^[4]. In addition, the scientific research to improve the performance of fuel cells has also dramatically increased. By 2024, a search on the web of science for “proton exchange membrane fuel cells” revealed that the number of papers and review articles published in the past 20 years has shown an increasing trend year by year (Fig. 1d). Among them, the number of papers about bipolar plates also increases markedly each year (Fig. 1e). In particular, more than 250 articles and reviews were published in 2022. It can be seen that development of high-performance bipolar plates has attracted extensive attention from researchers.

Classified by different types of graphite raw materials, bipolar plates can be divided into metal bipolar plates, graphite bipolar plates and composite bipolar plates^[33] (Fig. 1f and Table 1). The metal bi-

polar plate is generally made of stainless steel, titanium and other metals, and is prepared by metal forging^[34], stamping, forming and surface coating^[35–43]. It has the characteristics of relatively low cost, high mechanical strength, easy processing and low gas permeability. In addition, due to the excellent strength and ductility of metal materials, the bipolar plates can be molded thinner, assembling fuel cell stack with higher volume power density. Currently, Toyota and Hyundai all use metal bipolar plates in their fuel cells. However, PEMFCs usually work in complex working conditions (such as irregular voltage changes) and corrosive electrolyte environment (acidic, humid and 60–80 °C hot temperature). It is easy to cause the corrosion of metal bipolar plate, increasing its contact resistance^[44–50]. What is more serious is the dissolution of metal ions from the plate, which will poison the proton exchange membrane and catalyst, then reducing the performance of fuel cells. The coating can effectively protect the bipolar plate and extend its life. But it needs extra processing step and cost and to be further verified in the actual application.

Nonporous graphite plate is graphitized by

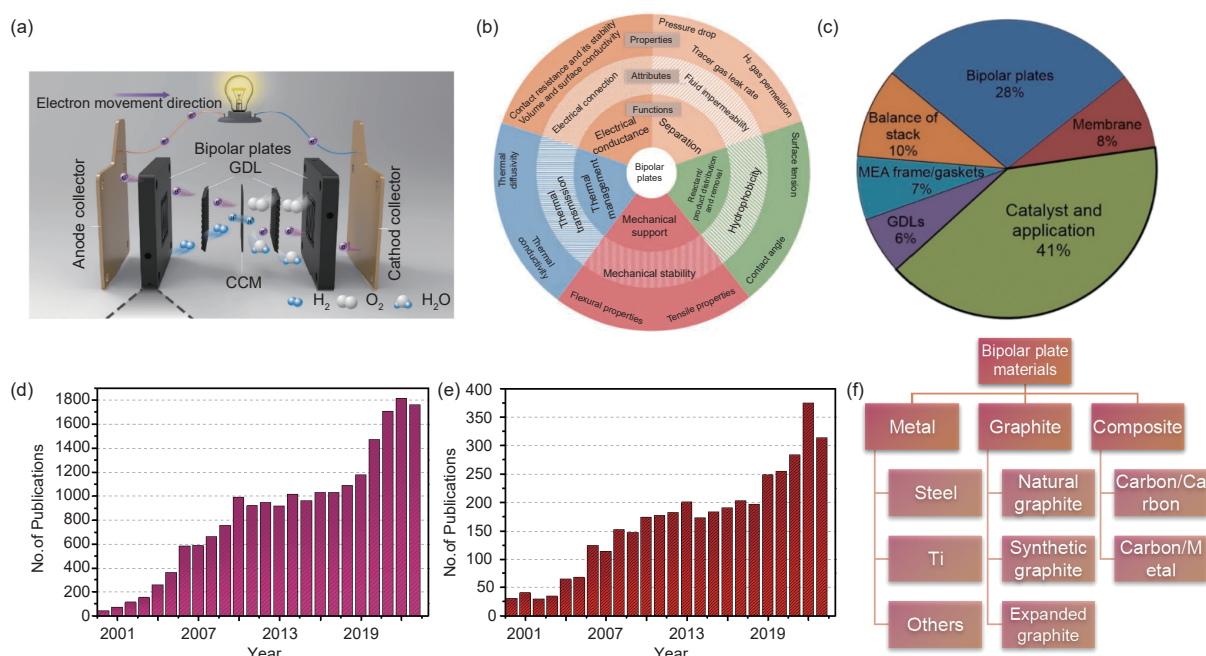


Fig. 1 (a) Schematic diagram of fuel cell^[23]. (b) Schematic diagram of functions of bipolar plate^[31]. (c) Cost breakdown of fuel cell at 500 000 units/year^[32]. The numbers of paper from 2000 to 2023: (d) proton exchange membrane fuel cells and (e) bipolar plates. (f) Bipolar plates classified by materials (Reprinted with permission)

Table 1 Advantages and disadvantages of different bipolar plate (BP) materials

Material	Advantages	Disadvantages
Metal	High thermal and electrical conductivity; superior mechanical properties; ease of fabrication; low cost; low gas permeability; excellent structural durability against shock and vibration; can form ultra-thin BP.	Prone to corrosion (membrane and catalysts poisoning, passive film formation).
Non-porous graphite	Excellent corrosion resistance; high thermal and electrical conductivity; stable chemical properties; mature manufacture technology.	Poor mechanical properties (brittleness); high weight and volume; poor machinability; high processing cost.
Carbon/ metal	As for uncoated metals.	Extra processing step and cost.
Composite	Graphite composite Excellent corrosion resistance; high thermal and electrical conductivity; stable chemical properties; lightweight; ease of forming flow fields.	Lower conductivity than metals and graphite; Less impermeable to gas than metals.

moulding the plate consisting of pitch and graphite powder, then engraving flow channel on it. This type of plate can change the flow channel easily, is the main plate of the current market. It also has the advantages of excellent corrosion resistance, high thermal and electrical conductivity, stable chemical properties and mature manufacture technology. However, the poor mechanical properties (brittleness) and high weight and volume are not good for its application. Additionally, the graphitization is time-consuming and energy consuming, and the high processing cost of engraving flow channel owing to its poor machinability also greatly restricts the production capacity of the plate.

Besides, due to the shortcomings of both metal and nonporous graphite bipolar plates, the composite bipolar plate comes into the vision of researchers. The composite bipolar plate^[51] can be divided into carbon-metal composite bipolar plate and composited graphite bipolar plate. Composited graphite bipolar plate is a pressed plate that consists of graphite as the main filler and polymer as binder^[52–56]. Due to its lightweight, ease of forming flow fields, the excellent chemical stability, electrical and thermal conductivity, it has been widely concerned and applied in the development of fuel cells. However, the porosity and brittleness of the graphite material make the strength and gas permeability of the bipolar plate poor^[57], so it is necessary to be impregnated by pitch or resin to in-

crease its mechanical properties and lower its gas permeability. Exquisite graphite structure design and resin matrix tailoring is crucial because the mechanism of gas permeation in composite bipolar plates (BPs) is determined by intrinsic properties of the graphite and matrix^[58–65]. Selecting right filler like high aspect ratio can reduce the permeation of hydrogen and oxygen gases. Strengthening the coordination between graphite powder and resin matrix, and improved manufacturing process both can help solve these problems.

Here, we focus on the bipolar plates with graphite as the main filler, including nonporous graphite plates and graphite polymer composite plates, which are summarized as graphite-based bipolar plates. Graphite polymer composite plate is generally a composite material prepared by molding graphite material with polymer, conductive filler and plugging agent, etc. It has been widely concerned by scientific research and industry at present.

The different types of graphite for the preparation of bipolar plate provide more filler options for high performance bipolar plate. Generally, according to the different types of graphite used, graphite bipolar plates can be roughly divided into three categories: natural graphite bipolar plates, synthetic graphite bipolar plates and modified graphite bipolar plates^[66]. Natural graphite exhibits good crystallization, superior electrical and thermal conductivity, making it an excellent filler. Artificial graphite is prepared by the

carbonaceous precursors after the graphitization at 2800 °C. Compared with natural graphite, the crystallinity of it is slightly worse, but it is not limited to graphite deposits, and is an important supplement to the graphite market. Modified graphite here is expanded graphite^[67], a worm-like porous graphite material prepared by oxidizing and peeling the crystallized graphite with modified Hummers' method^[68]. Due to the interlacing of the stripped graphite sheets, the expanded graphite not only has the excellent properties of the graphite material itself, but also increases the flexibility, which is helpful to improve the mechanical property of bipolar plate.

2 Functions and developed goals of bipolar plate

The bipolar plate plays an important role in the fuel cell stack, so the high performance of the bipolar plate is the key to the high performance of fuel cell. It is very important to evaluate the performance of the bipolar plate. The U.S. Department of Energy is a pioneer in the development of fuel cells, and has also put forward goals for the development of bipolar plates. In 2017, the Driving Research and Innovation for Vehicle Efficiency and Energy Sustainability in the United States also compiled a roadmap for the development of fuel cells and set new goals for the development of bipolar plates (Fig. 2^[69]).

One of the important functions of the bipolar plate is to transport the reaction gas, to conduct the current and water generated by the reaction, and to release the heat. Therefore, first of all, the plate materi-

al must have good electrical conductivity. Graphite has good electrical conductivity, but the polymer has poor electrical conductivity. How to strike a balance between the graphite filler and the polymer is the key to prepare the highly conductive graphite bipolar plate. The targets of graphite plate in 2025 need to have the electrical conductivity greater than 100 S/cm and the area specific resistance less than 0.01 $\Omega \text{ cm}^2$, and the conductivity and areal-specific resistance need to be tested with a four-probe instrument according to relevant standards^[12]. In addition, the surface of the bipolar plate needs to be processed as a channel for gas and water transmission, so the plate material also needs to have a certain degree of machinability. As we know, graphite is a brittle material, and improving its mechanical strength is vital step for its application. In 2025, the flexural strength (three-point flexure method^[12]) of the bipolar plate will reach 40 MPa, so as to better meet the need during its use. The inherent porosity of graphite also makes the gas permeability of the graphite plate weakness. The bipolar plate needs to separate oxygen and hydrogen in each chamber, so it must have low H_2 permeability (gas permeability tester^[12]) about $2 \times 10^{-6} \text{ cm}^3 \text{ s}^{-1} \text{ cm}^{-2} \text{ Pa}^{-1}$. On the other hand, with the dramatic increase of the power density of the fuel cell stack, the current density of plate keeps increasing, resulting in a large amount of heat generation. Therefore, the bipolar plate also needs to have a high thermal conductivity and a low coefficient of thermal expansion (infrared thermal imaging camera^[12]). And the working environment of fuel cell is acidic, so the bipolar plate

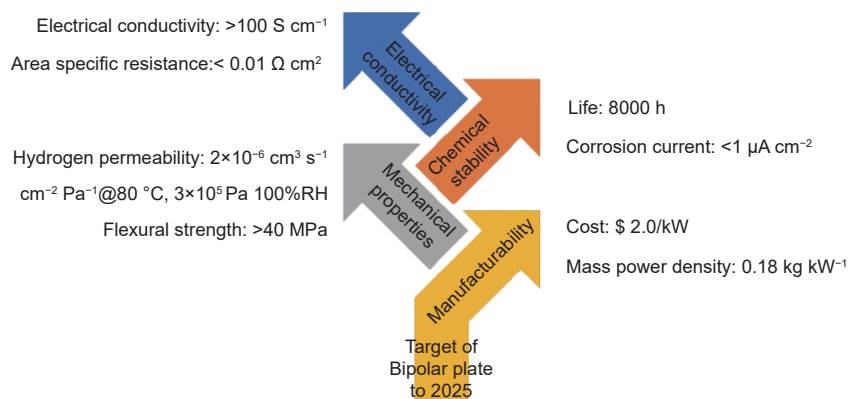


Fig. 2 The goals of bipolar plate in 2025 set by the United States Driving Research and Innovation^[69] (Reprinted with permission)

should have good chemical and electrochemical corrosion resistance to extend its life. The proposed target for corrosion current (utilization of constant potential and potentiodynamic polarization tests) of bipolar plate is less than $1 \mu\text{A cm}^{-2}$, so that the plate life can be longer than 8000 h. Finally, to achieve larger-scale application, it requires the bipolar plate to be thinner and lighter, increasing the mass/volume power density to 0.18 kg/kW by 2025, and the cost needs to be continuously reduced to \$2 /kW.

3 Factors affecting the performance of graphite-based bipolar plate

In general, the performance of the graphite bipolar plate is affected by 2 aspects, which are raw material and the synthesis process (Fig. 3). On the one hand, the material of graphite bipolar plate can be roughly divided into 3 parts, which are graphite filler, also known as primary filler, polymer matrix and reinforcement filler, also known as secondary filler. The electrical and thermal conductivity of graphite plate is determined by the purity and electrical/thermal conductivity of graphite raw material. The size, morphology and structure of fillers greatly affect the overall performance of graphite bipolar plate. Polymer is the binder of fillers, generally divided into thermoplastic and thermosetting resin. It can seal the plate to increase the mechanical property and gas impermeability. The reinforcement filler can enhance some properties of the graphite bipolar plate, like conductivity,

mechanical property and so on. From the aspect of raw materials, optimizing its structure can greatly increase the performance of graphite bipolar plate.

On the other hand, the processing is another major factor affecting the manufacturing of graphite plate, which can be roughly divided into three parts: the premixed process of raw materials, the molding process and the impregnation process. Premixing is the pre-step of molding process, and uniform mixing of raw materials is the basis of high performance of plate. Researchers need to pay more attention to the technical details and equipment. Molding process, one of the most critical steps, directly affects the thickness and flow channel processing of graphite plate. The impregnation is to solve the problem of insufficient sealing of the graphite plate after forming. The impregnated polymer can fill the gaps in the graphite plate and further enhance the mechanical strength of it. Through the optimization of raw materials and processes, bipolar plates with better performance can be prepared. The performances of graphite-based bipolar plates in published papers are listed in Table 2. The subsequent part of this review will detail the research progress of graphite-based bipolar plates. Finally, the author will put forward some insights for the subsequent research and development of graphite-based bipolar plates.

3.1 Materials and optimization

Graphite, a kind of non-metallic material, is also a crystal with a layered structure composed of many layers of sp^2 bonded carbon atoms arranged in a

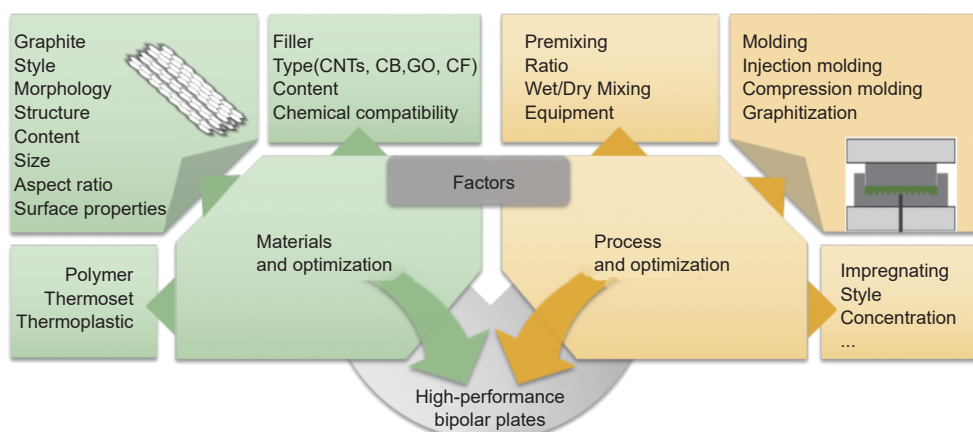


Fig. 3 Factors affecting the performance of graphite-based bipolar plates

Table 2 The performance of graphite-based bipolar plates in published papers

Graphite/%	Matrix/%	Filler/%	Production method	Density/ (g cm ⁻³)	Through-plane electrical conductivity / (S cm ⁻¹)	In-plane electrical conductivity / (S cm ⁻¹)	Flexural strength/ MPa	Ref.
G (82.5)	PP (15)	CB (2.5)	Twin-screw extrusion process	—	25.7	—	34.8	[70]
G (80)	Epoxy (20)	—	—	—	—	50.0	19.0	[71]
G (34)	PVDF (60)	CB (6)	Compression molding	—	—	177.9	49.2	[72]
G (35)	PVDF (60)	MWCNTs (5)	Compression molding	—	—	161.6	25.0	[72]
G (76)	Epoxy resin-bisphenol A diglycidyl ether	—	Compression molding	1.60	—	221.0	52.0	[73]
30% NWCNTs and CNF	PP (70)	—	Compression molding	—	—	8.2	45.3	[74]
SG (81)	Epoxy resin (15)	GN (4)	Compression molding	—	—	266.7	40.1	[75]
G (74.5)	Benzoxazine resin (16)	CNTs (2) GO (7.5)	Compression molding	—	—	364.0	41.5	[76]
NG (50)	Polycarbonate (62)	CNT (3) CF (10)	Compression molding	—	12.8	8.3	—	[77]
Milled CF (65)	PP (30)	GN (5)	—	—	14.8	—	172.5	[78]
EG: NG (25:25)	Phenolic resin (50)	—	Compression molding	1.55	—	>100.0	49	[79]
SG (77)	PP/epoxy blend (15)	CB (8)	Compression molding	—	8.5	88.5	28.5	[80]
G (50)	PP (20)	CB (25)	Compression molding	1.64	—	158.32	29.86	[81]
EG(80)	Phenolic resin(20)	—	Compression molding	1.55	—	182.0	109.0	[11]
G (87)	Epoxy resin (11)	CB (2)	Compression molding	—	—	162.0	48.0	[82]

Note: G: graphite. PP: polypropylene. CB: carbon black. PVDF: polyvinylidene fluoride. MWCNTs: mutli-walls carbon tubes. SG: synthesised graphite. NG: natural graphite. CF: carbon fiber. EG: expanded graphite. GN: graphene nanosheets

hexagonal^[83–85]. The interlayer of graphite is bonded by van der waals force (Fig. 4a). Because of the layer crystallization, graphite shows a good electrical conductivity, making it attractive as filler. In addition, the excellent chemical stability of graphite material is another major advantage, which is the guarantee of long-life of fuel cell. However, the brittleness and porosity of graphite material make bipolar plate machinability and gas permeability as challenges, which also requires more efforts from researchers. In addition, due to the anisotropy of graphite structure, the difference between axial and radial conductivity of the bipolar plate is increased, which limits the further enhancement of the plate performance. All in all, the type, morphology, structure, size, aspect ratio, surface properties and content of graphite have a great impact on the performance of graphite bipolar plate^[86]. The main idea for preparing high-performance graphite plate is to optimize the graphite material. The following are a summary and analysis of the influence of graphite material properties on plate performance.

Generally speaking, graphite materials can be divided into natural graphite and artificial graphite. Natural graphite is from mine after grinding, grading, purification and surface treatment, and its crystallin-

ity formed naturally. Artificial graphite is preparation from carbon-rich precursor after graphitization, and its degree of crystallization formed by heat treatment at high temperature^[90]. Due to the difference formation way, they have different morphology and crystal structure, thus showing different physical and chemical properties. The crystallinity of natural graphite is more complete, so the conductivity and thermal conductivity of it are higher than that of artificial graphite. In terms of microstructure, there are a large number of voids in the crystalline structure of natural graphite, while the artificial graphite is compact and seamless. Saadat et al.^[91] found that natural graphite resin composites had 8% higher electrical conductivity than artificial graphite composites, and relatively little difference in flexure strength. The authors speculated the reason was that the surface energy difference between natural graphite and polymer was larger, thus achieving the high electrical conductivity of graphite bipolar plates (221 S cm⁻¹) and high flexure strength (52 MPa). In terms of the morphology of graphite, it can be roughly divided into flake graphite, spherical graphite, expanded graphite, etc. (Fig. 4b-g). Because of its naturally layered crystalline structure, natural graphite usually is a flake (Fig. 4b). Artificial

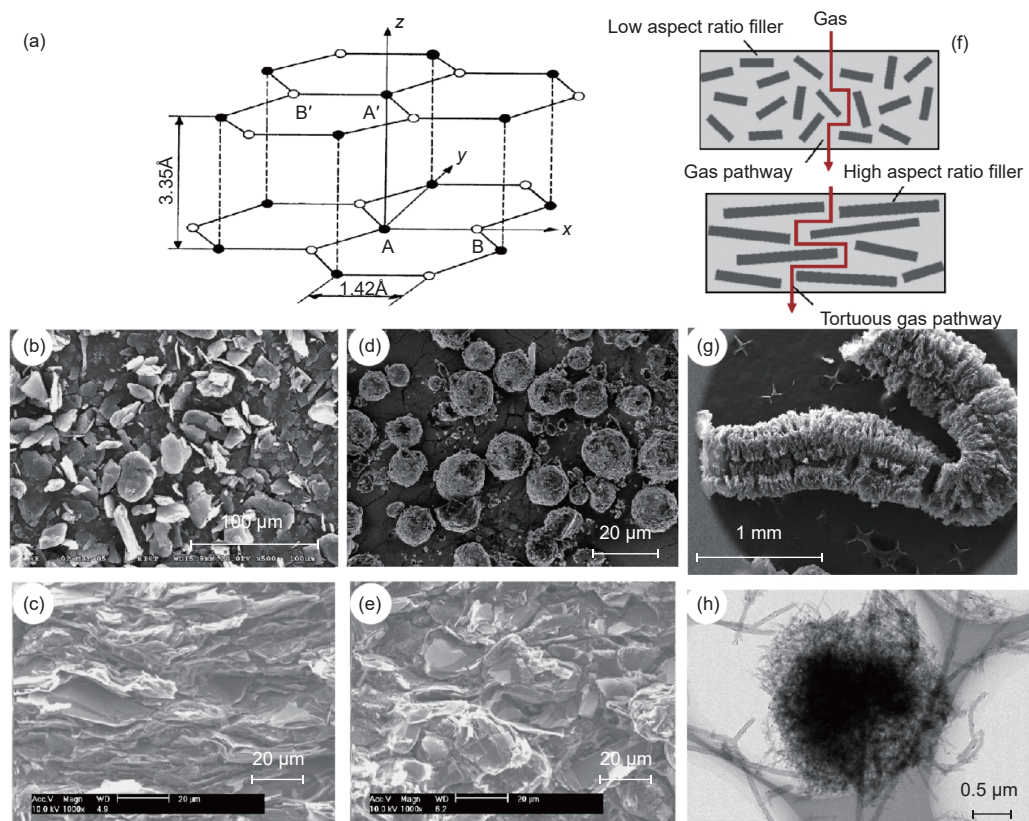


Fig. 4 (a) Graphite structure^[68]. SEM images of (b) flake graphite, (c) cross-section of graphite plate prepared by flake graphite, (d) spherical graphite^[87] and (e) cross-section of graphite plates prepared with spherical graphite^[88]. (f) Model diagram of influence of aspect ratio of graphite material on plate performance^[57]. (g) SEM image of expanded graphite-graphite worm^[68]. (h) TEM images of graphite microspheres woven by CNTs^[89]

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graphite morphology can also be flake or spherical (Fig. 4d). The effect of morphology and size of graphite to the performance of graphite bipolar plates has been studied. Flake graphite is more prone to orientation in the process of compression molding (Fig. 4c), resulting in better electrical and thermal conductivity of graphite plate composite along the orientation direction, but poor electrical conductivity in the vertical orientation directions. To some extent, the further improvement of graphite plate performance is restricted. Spherical graphite is not easily oriented during compression molding, so the difference between horizontal and vertical properties of the graphite plate is reduced (Fig. 4e). On the other hand, the difference of surface area will affect the binding force between the graphite material and the polymer, thus affecting the flexure strength of the graphite plate. Heo et al.^[88] studied the influence of the particle size and morphology of graphite particles on the GBP. When the dens-

ity is basically the same, the bipolar plate made of flake graphite particles is more compact and its electrical conductivity is higher than that of the plate made of spherical graphite. And the graphite with smaller particle size will lead to higher interface consistency between the graphite particle and phenolic resin, finally bringing higher flexure strength of the BP. The influence of interfacial correlation is also reflected in the change of particle shape. Flake graphite has a larger specific surface area, so the BP prepared by flake graphite has higher flexure strength. Some studies have proved that the increase of graphite size worsens the physical properties of the BP, resulting in the deterioration of the gas impermeability. Meanwhile, the conductivity of the BP also decreases with the decrease of the particle size of graphite, which makes it difficult to form an effective conductive network between graphite particles (Fig. 4f).

Suherman et al.^[71] studied the effect of graphite

particle size on the electrical conductivity of graphite-phenolic composites. The author used 80% graphite with particle size of 150 μm and 20% phenolic resin to prepare the graphite composite with electrical conductivity up to 23 S/cm. When 10% graphite with particle size of 75 μm was substituted for large graphite particle size, conductivity can be increased to 32 S/cm. In order to further investigate the effect of different graphite on the properties of bipolar plates, Kang et al.^[87] systematically studied the effects of graphite source, morphology and size on the properties of bipolar plates, and found that flake natural graphite showed the best electrochemical and mechanical properties due to its high crystallinity and aspect ratio. Synthetic graphite has higher surface energy than natural graphite, so it can be better wetted by the resin and show better bending strength. Therefore, the author believes that natural graphite can be used as an excellent primary filler and synthetic graphite can be used as a better secondary filler. At the same time, the authors found that large graphite particles can improve the overall electrical conductivity of the plate, but can reduce the bending strength of the plate. Therefore, the size of conductive filler should be selected reasonably according to the requirements of electrical conductivity and mechanical strength of the bipolar plate in the fuel cell stack. In addition, modification of graphite is another idea to improve the performance of bipolar plates. Lee et al.^[92] prepared large-particle graphite modified with fluorinated ethylene propylene powder by ball milling, mixed well-dispersed small-particle graphite and the modified large-particle graphite, and investigated the influence of dispersion and filling density. By modulating the electrical transmission path between large graphite particles with well-dispersed ethylene propylene fluoride, the orientation and dispersion of graphite particles in the matrix led to the improvement of electrical conductivity and mechanical properties. It provides a robust and scalable strategy to achieve high performance and large area bipolar plate.

Expanded graphite^[93] is a degree of separation of the carbon layers in the graphite, which may or may

not occur in adjacent carbon layers, so that the graphite remains intact, like a worm (Fig. 4g). If the carbon layer is completely separated and the carbon layer is less than 10 layers, it can become graphene material. The peeled graphite worms exhibit different physical and chemical properties from conventional graphite, especially increased flexibility. Its unique worm porous structure can be used to strengthen the electrical and thermal conductivity of the bipolar plate, and the excellent wettability of the resin material makes it easier to be manufactured. Expanded graphite has a larger surface area, and the worm structure is flexible. Therefore, when used as the filler bipolar plate, it shows better flexure strength. Masand et al.^[79] combined expanded graphite, natural graphite and carbon black as filler to prepare high performance composite graphite plate, because the large surface area of expanded graphite could make the resin better wet the graphite filler. The addition of natural graphite helped to increase the contact between expanded graphite and resin. In addition, several novel graphite structures have been designed. In 2021, Wang et al.^[89] reported the woven graphitized microsphere architected by CNTs (Fig. 4h). The structure has excellent electrical conductivity, while the woven structure of carbon nanotubes shows a small density, which is significance for the lightweight of fuel cell bipolar plates. In addition, there are a lot of structural innovation of graphite materials, which can enrich the graphite filler of bipolar plate materials, and help us to develop graphite bipolar plates with better performance. In this field, researchers still need to make a lot of efforts.

Reinforcement materials are added to enhance a certain aspect performance of composite bipolar plate. They are generally carbon-based materials, such as CB, graphene, CNTs and CF. The addition of these nanomaterials helps graphite particles form a conductive pathway in the resin matrix and improves the overall electrical conductivity of the plate (Fig. 5a). At the same time, the addition of filler with high aspect ratio can increase the permeation path of gas molecules and prevents the permeation of gas to the com-

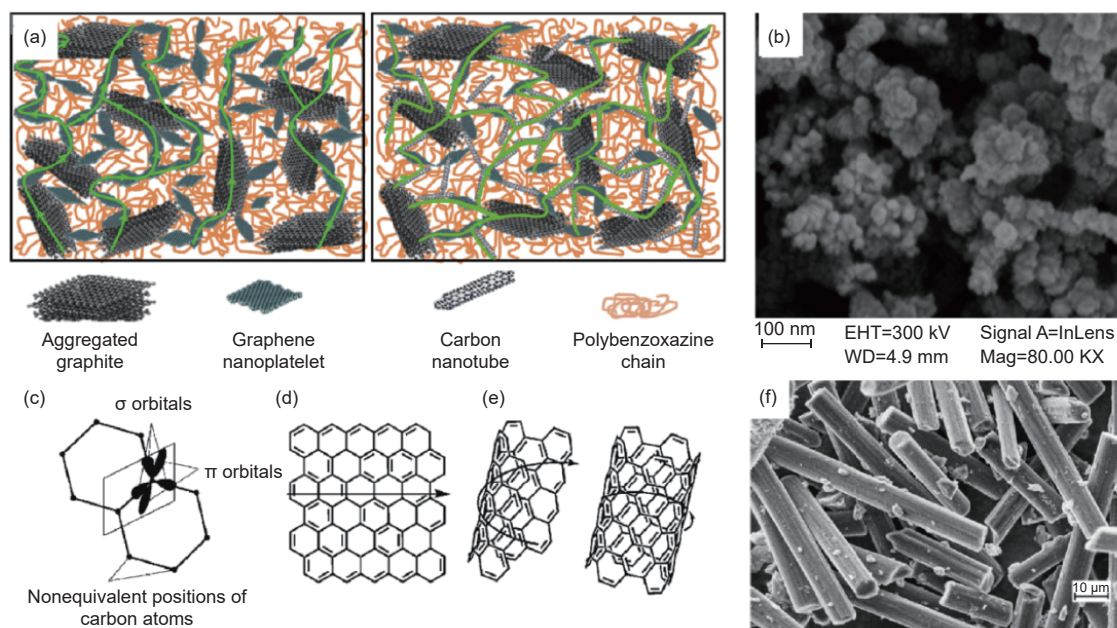


Fig. 5 (a) Schematic of the conductive network in composite^[94]. (b) SEM image of carbon black^[95]. (c) Orbital structure of graphene, (d) graphene structure, and (e) carbon nanotube structure. (f) SEM image of carbon fiber^[78] (Reprinted with permission)

posite plate. Graphene, CNTs and CF are considered as promising gas barrier materials due to their high aspect ratio. In addition, these fillers also have better mechanical strength, which can enhance the strength of the bipolar plate. Rich functional groups of these fillers also increase the interface bonding force between the graphite powder and the resin matrix, further strengthening the flexural strength of the bipolar plate. All in all, these nano-fillers are very excellent multi-functional fillers.

Buhler et al.^[96] evaluated electrochemical and mechanical properties of all carbon nano-materials which can be fillers of BP, such as graphite, CB (Fig. 5b), CNTs (Fig. 5e), CF (Fig. 5f) and EG. The addition of carbon black can increase the conductivity of GBP but reduce the flexure strength. The authors concluded that 15% addition of CNTs is the optimal value for the GBP through experiments. The addition of CF does not significantly increase the electrical conductivity and mechanical properties of the GBP, which was attributed to the poor compatibility between the CF and the non-polar PP matrix. But the addition of 1.5% silane coupling agent could increase the wettability of the filler and enhance the electrical conductivity and mechanical properties.

Owing to the nano-size, CB can more significantly

improve the chemical performance of the BP. Lim et al.^[97] reduced the electrochemical impedance of the composite BP through spraying the conductive particles (graphite and carbon black) in the epoxy matrix. The impedance of BP with 10% (mass fraction) addition of CB is only half of that of the composite BP without CB. However, the addition of too many nanoparticles can reduce other properties of the BP, and the addition of CB above 10% (volume fraction) will reduce the conductivity and flexure strength^[71]. Witpathomwong et al.^[76] investigated the effect of CNTs content on the properties of polybenzoxazine graphite/graphene composites. CNTs can be randomly oriented in composites and effectively bind to graphite and graphene. Thus, the thermal conductivity and electrical conductivity of composites are greatly improved. The author found that the thermal conductivity of the composite plate with 2% CNTs reached 21.3 W/mK, which was 44 times higher than that without CNTs. The conductivity also reached 364 S/cm and the flexure strength reached 41.5 MPa.

Tariq et al.^[70] prepared a graphite-PP based BP through the twin screw extrusion technique, and explored the influence of multiple fillers (multi-walled CNTs and CB) on the graphite-PP bipolar plate system. It is found that the conductivity of the multiple

filler system is 127% of that of the single-filler system, and the flexural strength of the multiple filler system is also enhanced. Ramirez-Herrera et al.^[74] also used two different fillers (multi-walled CNTs and carbon nanofibers) to enhance the performance of the composite BP. Compared with using either filler alone, the filling of 30% hybrid filler significantly enhanced the performance of the composite BP. Zambrzycki et al.^[75] reported the effects of secondary carbon fillers on the electrical, thermal and mechanical properties of epoxy resin/graphite composite plates. They compared the effects of 2 low-cost fillers (graphene nanosheets and CB) on the overall performance of composite plates. The authors found that graphene nanosheets with high specific surface area, large aspect ratio, and nanosized can significantly improve the performance of composite plates, while the addition of nano-spherical CB has limited improvement effect on composite plates. Naji et al.^[77] used three fillers (3% CNTs, 10% CF, 50% graphite) to prepare composite plates with improved properties compared with single filler and 2 fillers. It can be seen that the combination of multiple carbon fillers can better form a conductive pathway in the polymer matrix and further improve the conductive properties. However, each filler has its own advantages and disadvantages, how to make better coordination between fillers to prepare composite BP with enhanced performance still needs more efforts of researchers.

On the other hand, the orientation of the filler is also critical to the performance of the BP. Radzuan et al.^[78] found that the orientation of filler had a great influence on the conductivity and mechanical properties of composite BP. The authors enhanced the conductivity and flexural strength of the prepared composite BP by making fiber orientation through extrusion process before molding. This study revealed that oriented fibers can enhance the performance of composite BP, but the vertically conductivity of oriented plates will reduce. The carbon-based material has a strong anisotropy, so the orientation of the filler can enhance the electrical/thermal conductivity and mechanical properties in the orientation direction, but the

vertical orientation direction does not change greatly. In this regard, more research is needed to verify the balance and make it move towards the direction of comprehensive improvement of composite plate performance.

As an important matrix material for BP, polymer helps to bond brittle graphite powder and improves the gas impermeability of the plate. It is a key material that affects the flexure strength, gas impermeability and electrical and thermal conductivity of the BP. Different polymers have different crystal structures, morphology and viscosity, which are all key parameters affecting the overall properties of composite BP. In addition, the wettability between the polymer and the filler also greatly affects the performance of the composite BP. When the surface energy between the polymer and the filler is similar, the polymer can better wet the surface of the filler. Resin, the most commonly used matrix material of BP, can be generally divided into thermoplastic resin and thermosetting resin. Thermoplastic resin is a polymer with a long chain molecular structure, and the molecular chain of thermosetting resin is generally a body type network structure. Thermoplastic resins cure to a certain mechanical strength before they are removed from the mold, while thermosetting resins cure quickly through pressure and heat, so they can be removed from the mold without cooling, speeding up the production. However, post-curing reactions are sometimes required to eliminate residual monomers that may poison the membrane electrode assembly and reduce its ionic conductivity.

Thermoplastic resin is suitable for a wide variety of applications due to its inherent properties such as low density, easy-processing and recyclability. Many authors have also investigated the effects of using different thermoplastic resins, the most common being polypropylene (PP). It has advantages such as low cost, good processing conditions and mechanical properties, but the lack of polar functional groups leads to poor dispersion of graphite powder in the resin matrix, which further leads to the difficulty of forming an effective conductive pathway for graphite

particles in the matrix and the difficulty of further improving the conductive properties. Some researchers have solved this problem by grafting polar molecules onto PP monomers. Dhakate et al.^[98] also proposed that polymers with polar groups were more conducive to wetting the filler, making it form a conductive path better and enhancing the conductivity of composite materials. The high mechanical strength and thermal stability of polyphenylene sulfide (PPS) make it an excellent polymer matrix for the preparation of high-content filler of BP. PPS is a kind of semi-crystalline polymer, and its excellent high temperature resistance and recyclability make it very attractive. However, there are still few studies on the corrosion resistance of PPS. In this respect, further research is needed to promote its further application. Polyvinylidene fluoride (PVDF) is a resin with good barrier properties, chemical inertia, good mechanical properties and moisture resistance. Hu et al.^[72] prepared a GBP with low filler and collaborative separation structure by moulding 34% graphite-60%PVDF-6% CB, achieving a high conductivity of 177.8 S/cm. The high flexure strength of 49.16 MPa provides an important idea for the application of PVDF in GBP.

The biggest characteristics of thermosetting resin is strong, tough and low viscosity. It can be used to prepare the plate with large filling volume, and it can be molded without additional process. However, the disadvantage of thermosetting resin is not recyclable and cannot be produced by traditional injection molding and sheet extrusion process. Various types of thermosetting resins have been studied in the literature as possible matrix for composite BP, and commonly used resins include epoxy, phenolic and vinyl resins. Epoxy resin is a polymer containing 2 or more epoxy groups in its molecular structure. It was first put into production in the United States in 1974. In the 1950s, a large number of new varieties emerged, and great progress was made in its application. Phenolic resin has good mechanical properties and thermal stability, curing between 60–140 °C. At the same time, it has polarity and double bond structure, and can be well compatible with graphite filler, so that it is widely

used in the manufacture of BP. Vinyl resin can be seen as a cross between polyester and epoxy, with excellent impedance and corrosion resistance. While simultaneously combining the characteristics of light, strong, tough and low cost, it also has the prospect of large-scale use.

The characteristics of each resin are distinctive. Alo et al.^[80] mixed different resins to prepare plates with better performance. The authors used graphite and carbon black as the hybrid filler, and use PE, epoxy (EP) and PE/EP respectively as the matrix materials of BP. It is found that the non-polar semi-crystalline polymer PE with obvious amorphous region is easy to form conductive network when it is filled with graphite and CB with relatively small size, because these fillers are more likely to aggregate in the amorphous region of the matrix rather than in the bulk matrix material. However, due to lack of polar groups in PE, the interaction between PE and the filler is insufficient, which is not conducive to the uniform dispersion of the filler in the matrix. PP and epoxy resin blends have also been tested as matrix material to synthesize GBP. PP/epoxy blends have a co-continuous structure, which is conducive to the formation of continuous conductive network of filler and the improvement of electrical conductivity. The maximum in-plane and through-plane conductivities are 90.34 and 9.34 S cm⁻¹, respectively. It can be seen that the resin blend can overcome the performance defects of a single resin and make the matrix material play a better role^[80].

3.2 Processing technology and optimization

The dispersion of filler and polymer matrix determines the electrical conductivity, thermal conductivity and mechanical properties of composite BP. The poorly dispersed graphite particles or reinforcing materials cannot form effective conduction electrical and thermal paths in the polymer matrix, so more effort should be paid to the premixing process of raw materials. Chaiwan et al.^[82] studied the effects of wet and dry dispersed CNTs on the properties of phenolic resin matrix with high graphite content. It was found that the composite with high filler content had better dis-

persion effect by using dry method, and the prepared composite showed higher electrical conductivity and mechanical strength, which also met the requirements of the United States Department of Energy for BP. Roncaglia et al.^[82] found that the method of wet mixing for the preparation of graphite epoxy resin composites was far superior to the dry mixing process. The authors believed that in the presence of solvent, the filler and resin could be dispersed more uniformly, which was more conducive to the electrochemical contact between the fillers. There is no unified conclusion on the dispersion of filler and polymer in the literature. The exploration of premixed technology needs to be further strengthened to guide the processing and manufacturing of BP.

The easy to scale manufacturing molding methods for plates with flow channels are including compression molding and injection-compression (Fig. 6). Compression molding is an important method to process most composite materials. It is a process in which the slurry is put into the mold cavity at the molding temperature, and then the mold is closed and pressurized to make it formed and cured. During the molding process, the resin composite needs to have good compatibility, fluidity and appropriate viscosity so that the resin can better bond to the graphite filler and fill the whole cavity, forming a uniform composite material. The injection molded GBP is to send a certain proportion of graphite and resin mixture from the hopper of the injection molding machine, and the mixture after heating and melting is injected into the closed mold through the nozzle under the pressure. After cooling and setting, the product is released. The production efficiency of injection molding is very high, making it suitable for mass production. It can realize mechanization and automatic production, and the product has accurate size and shape. However, in order to ensure the electrical conductivity of the BP, the proportion of graphite must be sufficient, so the proportion of resin must be relatively reduced, which leads the flow capacity of the injection material worse, and it is difficult to ensure the uniformity of the BP. It is necessary to increase the amount of resin to increase the fluidity of

the composite mixture, but it will sacrifice the electrochemical performance of the BP. Thus, the injection molding in the manufacturing process of BP has not been widely promoted. Compared with injection molding, compression molding reduces demand of the fluidity of mixture and can use a resin with lower viscosity. At the same time, the process is simple and mature, which has a good prospect in the large-scale preparation of GBP.

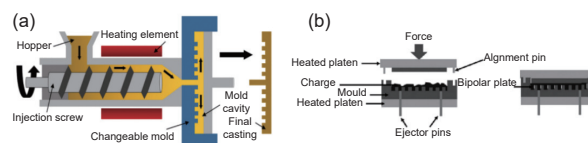


Fig. 6 Schematic of (a) injection-compression and (b) compression molding^[80,99] (Reprinted with permission)

In the manufacturing process of BP, impregnation process is generally used in the preparation of nonporous graphite plates and flexible graphite plates. In the manufacturing process of nonporous graphite plate, it is necessary to repeatedly impregnate with pitch until the air tightness meets the requirements. Flexible graphite plate is pressed into expanding graphite, and further impregnated with resin to enhance its mechanical strength and strengthen the gas impermeability. Fan et al.^[100] prepared the expanded graphite resin composite BP by hot pressing and vacuum impregnation process, and studied the influence of the impregnated resin concentration on the electrochemical and mechanical properties of the plate. When the concentration of water-soluble phenolic resin was 25%, authors found that the BP showed better mechanical properties and resistivity, and the flexure strength is 86.95 MPa and the resistivity is 8.24 mΩ·cm². The impregnated flexible graphite plate has been widely used in industry due to its simple process and excellent performance, gradually seizing the market share occupied by the nonporous graphite plate. However, processes (such as resin curing process and cleaning) and the problems (such as leaching at the working condition of fuel cell and failure at low temperature) need more efforts to solve. The continuous adjustment and optimization of precision industrial equipment with process parameters may be helpful

to solve these problems.

4 Summary and outlook

Graphite-based bipolar plates (GBPs) are promising for PEMFCs due to high conductivity and corrosion resistance, but conventional nonporous GBPs face hurdles in further thinning and time-consuming channel machining. Graphite-polymer composite BPs via compression molding offer a scalable alternative, yet their performance requires further optimization of materials and processes.

Graphite is the main material of GBP, whose basic properties determine the performance of BP. For graphite fillers, key parameters include type, size, aspect ratio, and morphology. Artificial graphite exhibits better electrical conductivity than synthetic graphite, while flake graphite tends to orient during molding, yielding excellent in-plane conductivity, whereas spherical graphite mitigates anisotropy. Larger graphite particles enhance conductivity but reduce surface area for resin bonding; higher aspect ratios improve mechanical performance and gas resistance. Expanded graphite, with its worm-like structure and abundant functional groups, offers flexibility, improves compatibility with polymers, and can even be molded without resin, simplifying processing.

Carbon-based nanomaterials (carbon black, CNTs, carbon nanofibers, graphene) serve as secondary fillers to construct conductive networks and reinforce mechanical properties. Zero-dimensional spherical CB can greatly improve the electrical conductivity of the composite plate with a small amount addition, but too much carbon black will lead to the deterioration of mechanical properties. One-dimensional CNTs and CF also have extremely high electrical conductivity and strong mechanical properties. A large aspect ratio of them can significantly increase the mechanical properties, making them an excellent multifunctional filler. Two-dimensional graphene material is a star material in the scientific research field. And the sheet structure of it can very well strengthen the bonding force between graphite and polymer. Synergistic combinations of multiple fillers can achieve su-

perior performance at low contents, but underlying mechanisms require further study.

Resin can be used as the polymer matrix material to bond graphite particles to strengthen the gas impermeability and mechanical properties of GBP. Both thermosetting and thermoplastic resins, including blends, have been investigated. In addition, the wettability between the resin and the filler greatly affects the performance of GBP. When the surface energy between the resin and the filler is similar, the polymer can better wet the surface of the filler, which puts forward higher requirements for the optimization of the resin.

The optimization of manufacturing process is the key to the qualified preparation of large-scale BP. Premixing of synthetic materials is the basis for preparing BP with uniform performance. The choice of dry or wet process needs to be made according to the properties of specific raw materials. There are various molding methods and each has its own advantages and disadvantages. In order to prepare a complete shape of the plate during injection molding, it is necessary to increase the amount of resin to increase the fluidity of the material, but it will sacrifice the conductive property of the plate itself, which constitutes a pair of contradictions, and a lot of experiments are needed to master the balance. The molding process is simple and easy, and it has been widely studied and practiced in the forming of metal materials. There are a lot of research experiences to learn from, so it has a good prospect in the large-scale preparation of graphite base plate materials. Impregnation is a supplementary process for the lack of air tightness and mechanical properties of graphite plate. The main problems of impregnation process are the type, concentration, curing temperature, leaching of resin and the failure of resin after freezing. In this respect, it is also necessary to increase investment in the process of industrialization to explore.

In conclusion, by rationally designing graphite morphology, selecting appropriate fillers, ensuring resin wettability, and optimizing molding and impregnation, high-performance composite GBPs can be

achieved. These materials are particularly suitable for stationary power generation and moderate-power-density applications, and they are poised to meet the demands of the growing hydrogen economy.

References

- [1] Cui Z, Li B Y, Xiong H, et al. A review of ways to improve the performance of hard carbon anodes in low-temperature sodium-ion batteries[J]. *New carbon materials*, 2025, 40(6): 1246-1263.
- [2] Xiao F, Wang Y C, Wu Z P, et al. Recent advances in electrocatalysts for proton exchange membrane fuel cells and alkaline membrane fuel cells[J]. *Advanced materials*, 2021, 33(50): 2006292.
- [3] Cao B, Cui Z, Liu H, et al. Conventional carbon anodes for potassium-ion batteries: progress, challenges and prospects[J]. *New carbon materials*, 2025, 40: 717-737.
- [4] Wang Y, Ruiz Diaz D F, Chen K S, et al. Materials, technological status, and fundamentals of PEM fuel cells-a review[J]. *Materials today*, 2020, 32: 178-203.
- [5] Fan L, Tu Z, Chan S H. Recent development of hydrogen and fuel cell technologies: a review[J]. *Energy reports*, 2021, 7: 8421-8446.
- [6] Jiao K, Xuan J, Du Q, et al. Designing the next generation of proton-exchange membrane fuel cells[J]. *Nature*, 2021, 595(7867): 361-369.
- [7] Manoharan Y, Hosseini S E, Butler B, et al. Hydrogen fuel cell vehicles; current status and future prospect[J]. *Applied sciences*, 2019, 9(11): 2296.
- [8] Ioroi T, Siroma Z, Yamazaki S I, et al. Electrocatalysts for PEM fuel cells[J]. *Advanced energy materials*, 2018, 9(23): 1801284.
- [9] Okonkwo P C, Ige O O, Barhoumi E M, et al. Platinum degradation mechanisms in proton exchange membrane fuel cell (PEMFC) system: a review[J]. *International journal of hydrogen energy*, 2021, 46(29): 15850-15865.
- [10] Mo J, Steen S, Kang Z, et al. Study on corrosion migrations within catalyst-coated membranes of proton exchange membrane electrolyzer cells[J]. *International journal of hydrogen energy*, 2017, 42(44): 27343-27349.
- [11] Yao K, Adams D, Hao A, et al. Highly conductive and strong graphite-phenolic resin composite for bipolar plate applications[J]. *Energy fuels*, 2017, 31(12): 14320-14331.
- [12] Warner T E, Graf T. On the measurement of the electrical conductivity of graphite-polymer composite bipolar plates[J]. *Journal of power sources*, 2024, 594: 233871.
- [13] Qi Y, Cui J, Guo F, et al. Graphene/polyaniline waterborne composite coatings for metallic bipolar plates[J]. *Progress in organic coatings*, 2024, 197: 108829.
- [14] Hanapi I H, Kamarudin S K, Beygisangchin M, et al. Review of recent composite bipolar plate in system development[J]. *Journal of environmental chemical engineering*, 2024, 12(5): 113776.
- [15] Guo F, Li J. Facile production of graphene-based ternary composite coatings on metallic bipolar plates[J]. *Applied surface science*, 2024, 669: 160360.
- [16] Choe J, Woo Lim J. Conductive nanoparticle-embedded carbon composite bipolar plates for vanadium redox flow batteries[J]. *Composite structures*, 2024, 329: 117770.
- [17] Tariq M, Utkarsh, Syed N A, et al. Optimization of filler compositions of electrically conductive polypropylene composites for the manufacturing of bipolar plates[J]. *Polymers*, 2023, 15(14): 3076.
- [18] Roncaglia F, Mucci A, Romagnoli M, et al. Graphite-epoxy composites for bipolar plates: the gas permeability issue[J]. *International journal of hydrogen energy*, 2023, 48(54): 20935-20944.
- [19] Musse D, Lee D. Parametric study of pulsed nanosecond laser interaction with carbon-nanotube composite bipolar plate for PEMFCs[J]. *Scientific reports*, 2023, 13(1): 2048.
- [20] Gaudig M, Münchgesang W, Martins de Souza E Silva J, et al. Electromechanical and electrochemical properties of highly filled titanium composites for PEM bipolar plates[J]. *Applied physics a*, 2023, 129(2): 1-10.
- [21] Darıcık F, Topcu A, Aydın K, et al. Carbon nanotube (CNT) modified carbon fiber/epoxy composite plates for the PEM fuel cell bipolar plate application[J]. *International journal of hydrogen energy*, 2023, 48(3): 1090-1106.
- [22] Yao D, Zheng J, Jin L, et al. Effect of surface oxidation on the interfacial and mechanical properties in graphite/epoxy composites composite bipolar plates[J]. *Chinese chemical letters*, 2024, 35(11): 109382.
- [23] Yang L, Zhao T, Yang W, et al. Design and preparation of composite bipolar plates with synergistic conductive networks of graphite and metal foam[J]. *International journal of hydrogen energy*, 2024, 92: 165-173.
- [24] Yan G, Zhang X, Wei L, et al. Study on the effects of nano-copper addition on the properties of epoxy resin/graphite composite bipolar plates[J]. *International journal of hydrogen energy*, 2024, 69: 576-585.
- [25] Xu G, Du X, Chang L, et al. Effect of molding temperature on the properties of phenolic resin/carbon black/graphite composite bipolar plates[J]. *Journal of solid state electrochemistry*, 2024, 28(8): 2505-2513.
- [26] Wilberforce T, Thompson J, Olabi A G. Bipolar plate materials[M]//Encyclopedia of Smart Materials. Amsterdam: Elsevier, 2022: 273-278.
- [27] Bian H, Du Y, Ren Y, et al. One-step electrodeposition of polypyrrole/Ti₃C₂T_x MXene composite coating for 304SS bipolar plates in PEMFC[J]. *Surface and coatings technology*, 2023, 462: 129460.

- [28] Alo O A, Otunniyi I O, Sadiku E R. Processing methods for conductive polymer composite bipolar plates: effect on plate quality and performance[J]. *Fuel cells*, 2023, 23(2): 136-160.
- [29] Zhang X, Huang H, Fan J, et al. Gradient structured composite bipolar plates for proton exchange membrane fuel cells[J]. *Acs sustainable chemistry & engineering*, 2022, 10(48): 15846-15856.
- [30] Park K S, Lee M H, Woo J S, et al. Fluorinated ethylene-propylene/graphite composites reinforced with silicon carbide for the bipolar plates of fuel cells[J]. *International journal of hydrogen energy*, 2022, 47(6): 4090-4099.
- [31] Shaigan N, Yuan X Z, Girard F, et al. Standardized testing framework for quality control of fuel cell bipolar plates[J]. *Journal of power sources*, 2021, 482: 228972.
- [32] Saadat N, Dhakal H N, Tjong J, et al. Recent advances and future perspectives of carbon materials for fuel cell[J]. *Renewable and sustainable energy reviews*, 2021, 138: 110535.
- [33] Mathew C, Mohamed N S, Devanathan L S. A comprehensive review of current research on various materials used for developing composite bipolar plates in polymer electrolyte membrane fuel cells[J]. *Polymer composites*, 2022, 43: 4100-4114.
- [34] Modanloo V, Alimirzaloo V, Elyasi M. Manufacturing of titanium bipolar plates using warm stamping process[J]. *Arabian journal for science and engineering*, 2020, 45(11): 9661-9667.
- [35] Weng F B. Review on proton exchange membrane fuel cell's metallic bipolar plate fabrication challenges[J]. *International journal of electrochemical science*, 2022, 17: 2052.
- [36] Liu R, Jia Q, Zhang B, et al. Protective coatings for metal bipolar plates of fuel cells: a review[J]. *International journal of hydrogen energy*, 2022, 47(54): 22915-22937.
- [37] Liu G, Hou F, Peng S, et al. Process and challenges of stainless steel based bipolar plates for proton exchange membrane fuel cells[J]. *International journal of minerals, metallurgy and materials*, 2022, 29(5): 1099-1119.
- [38] Wu S, Yang W, Yan H, et al. A review of modified metal bipolar plates for proton exchange membrane fuel cells[J]. *International journal of hydrogen energy*, 2021, 46(12): 8672-8701.
- [39] Xu Z, Qiu D, Yi P, et al. Towards mass applications: a review on the challenges and developments in metallic bipolar plates for PEMFC[J]. *Progress in natural science: materials international*, 2020, 30(6): 815-824.
- [40] Ouyang C. Physical and electrochemical properties of Ni-P/TiN coated Ti for bipolar plates in PEMFCs[J]. *International journal of electrochemical science*, 2020, 15: 80-93.
- [41] Madadi F, Rezaeian A, Edris H, et al. Improving performance in PEMFC by applying different coatings to metallic bipolar plates[J]. *Materials chemistry and physics*, 2019, 238: 121911.
- [42] Asri N F, Husaini T, Sulong A B, et al. Coating of stainless steel and titanium bipolar plates for anticorrosion in PEMFC: a review[J]. *International journal of hydrogen energy*, 2017, 42(14): 9135-9148.
- [43] Yi P, Zhang D, Qiu D, et al. Carbon-based coatings for metallic bipolar plates used in proton exchange membrane fuel cells[J]. *International journal of hydrogen energy*, 2019, 44(13): 6813-6843.
- [44] Leng Y, Ming P, Yang D, et al. Stainless steel bipolar plates for proton exchange membrane fuel cells: materials, flow channel design and forming processes[J]. *Journal of power sources*, 2020, 451: 227783.
- [45] Asri N F, Teuku Husaini T H, Sulong A B, et al. Effects of the synthesis coating parameters for metal bipolar plates[J]. *Sains Malaysiana*, 2020, 49(12): 3179-3188.
- [46] Gao P, Xie Z, Ouyang C, et al. Electrochemical characteristics and interfacial contact resistance of Ni-P/TiN/PTFE coatings on Ti bipolar plates[J]. *Journal of solid state electrochemistry*, 2018, 22(7): 1971-1981.
- [47] Cunningham B D, Huang J, Baird D G. Review of materials and processing methods used in the production of bipolar plates for fuel cells[J]. *International materials reviews*, 2013, 52(1): 1-13.
- [48] Zhang D, Duan L, Guo L, et al. TiN-coated titanium as the bipolar plate for PEMFC by multi-arc ion plating[J]. *International journal of hydrogen energy*, 2011, 36(15): 9155-9161.
- [49] Yu H N, Lim J W, Suh J D, et al. A graphite-coated carbon fiber epoxy composite bipolar plate for polymer electrolyte membrane fuel cell[J]. *Journal of power sources*, 2011, 196(23): 9868-9875.
- [50] Zhang D, Duan L, Guo L, et al. Corrosion behavior of TiN-coated stainless steel as bipolar plate for proton exchange membrane fuel cell[J]. *International journal of hydrogen energy*, 2010, 35(8): 3721-3726.
- [51] Planes E, Flandin L, Alberola N. Polymer composites bipolar plates for PEMFCs[J]. *Energy procedia*, 2012, 20: 311-323.
- [52] Wu S D, Chiou A H. The study on a new method of preparing PMMA forming composite bipolar plate[J]. *Scientific reports*, 2021, 11(1): 8753.
- [53] Yeetsorn R, Prissanaroon-Ouajai W, Onyu K. Assessment of surface resistance reduction on polypyrrole-coated composite bipolar plates[J]. *Journal of polymer research*, 2020, 27(8): 229.
- [54] Hao W, Ma H, Sun G, et al. Magnesia phosphate cement composite bipolar plates for passive type direct methanol fuel cells[J]. *Energy*, 2019, 168: 80-87.
- [55] Park H J, Woo J S, Park S Y. Poly(phenylene sulfide)-graphite composites for bipolar plates with preferred morphological orientation[J]. *Korean journal of chemical engineering*, 2019, 36(12): 2133-2142.
- [56] Lee M H, Kim H Y, Kim J, et al. Influence of oxyfluorinated graphite on fluorinated ethylene-propylene composites as bipolar plates[J]. *Carbon letters*, 2019, 30(3): 345-352.
- [57] Jeong K I, Oh J, Song S A, et al. A review of composite bipolar plates in proton exchange membrane fuel cells: electrical

- properties and gas permeability[J]. *Composite structures*, 2021, 262: 113617.
- [58] Gao P, Xie Z, Wu X, et al. Development of Ti bipolar plates with carbon/PTFE/TiN composites coating for PEMFCs[J]. *International journal of hydrogen energy*, 2018, 43(45): 20947-20958.
- [59] Lee D, Choe J, Nam S, et al. Development of non-woven carbon felt composite bipolar plates using the soft layer method[J]. *Composite structures*, 2017, 160: 976-982.
- [60] Li W, Jing S, Wang S, et al. Experimental investigation of expanded graphite/phenolic resin composite bipolar plate[J]. *International journal of hydrogen energy*, 2016, 41(36): 16240-16246.
- [61] Nam S, Lee D, Lee D G, et al. Nano carbon/fluoroelastomer composite bipolar plate for a vanadium redox flow battery (VRFB)[J]. *Composite structures*, 2017, 159: 220-227.
- [62] Lee D, Lim J W, Lee D G. Cathode/anode integrated composite bipolar plate for high-temperature PEMFC[J]. *Composite structures*, 2017, 167: 144-151.
- [63] Liu Z, Wang B, Yu L. Preparation and surface modification of PVDF-carbon felt composite bipolar plates for vanadium flow battery[J]. *Journal of energy chemistry*, 2018, 27(5): 1369-1375.
- [64] Lee D, Lee D G. Carbon composite bipolar plate for high-temperature proton exchange membrane fuel cells (HT-PEMFCs)[J]. *Journal of power sources*, 2016, 327: 119-126.
- [65] Ghosh A, Verma A. Carbon-polymer composite bipolar plate for HT-PEMFC[J]. *Fuel cells*, 2014, 14(2): 259-265.
- [66] Wang D, Zhang J, Dong Y, et al. Progress on graphitic carbon materials for potassium-based energy storage[J]. *New carbon materials*, 2021, 36(3): 435-448.
- [67] An Y, Fei H, Zeng G, et al. Commercial expanded graphite as a low-cost, long-cycling life anode for potassium-ion batteries with conventional carbonate electrolyte[J]. *Journal of power sources*, 2018, 378: 66-72.
- [68] Chung D D L. A review of exfoliated graphite[J]. *Journal of materials science*, 2015, 51(1): 554-568.
- [69] Xiong K, Wu W, Wang S, et al. Modeling, design, materials and fabrication of bipolar plates for proton exchange membrane fuel cell: a review[J]. *Applied energy*, 2021, 301: 117443.
- [70] Tariq M, Utkarsh, Syed N A, et al. Synergistic enrichment of electrically conductive polypropylene-graphite composites for fuel cell bipolar plates[J]. *International journal of energy research*, 2022, 46(8): 10955-10964.
- [71] Suherman H, Dweiri R, Sulong A B, et al. Improvement of the electrical-mechanical performance of epoxy/graphite composites based on the effects of particle size and curing conditions[J]. *Polymers*, 2022, 14(3): 502.
- [72] Hu B, He G, Chang F, et al. Low filler and highly conductive composite bipolar plates with synergistic segregated structure for enhanced proton exchange membrane fuel cell performance[J]. *Energy*, 2022, 251: 123982.
- [73] Wenkai L, Zhiyong X, Haodong Z. Current status of research on composite bipolar plates for proton exchange membrane fuel cells (PEMFCs): nanofillers and structure optimization[J]. *Rsc advances*, 2024, 14: 7172-7194.
- [74] Ramírez-Herrera C A, Tellez-Cruz M M, Pérez-González J, et al. Enhanced mechanical properties and corrosion behavior of polypropylene/multi-walled carbon nanotubes/carbon nanofibers nanocomposites for application in bipolar plates of proton exchange membrane fuel cells[J]. *International journal of hydrogen energy*, 2021, 46(51): 26110-26125.
- [75] Zambrzycki M, Sokolowski K, Gubernat M, et al. Effect of secondary carbon nanofillers on the electrical, thermal, and mechanical properties of conductive hybrid composites based on epoxy resin and graphite[J]. *Materials*, 2021, 14(15): 4169.
- [76] Witpathomwong S, Okhawilai M, Jubsilp C, et al. Highly filled graphite/graphene/carbon nanotube in polybenzoxazine composites for bipolar plate in PEMFC[J]. *International journal of hydrogen energy*, 2020, 45(55): 30898-30910.
- [77] Naji A, Krause B, Pötschke P, et al. Hybrid conductive filler/polycarbonate composites with enhanced electrical and thermal conductivities for bipolar plate applications[J]. *Polymer composites*, 2018, 40(8): 3189-3198.
- [78] Radzuan N A M, Sulong A B, Husaini T, et al. Fabrication of multi-filler MCF/MWCNT/SG-based bipolar plates[J]. *Ceramics international*, 2019, 45(6): 7413-7418.
- [79] Masand A, Borah M, Pathak A K, et al. Effect of filler content on the properties of expanded-graphite-based composite bipolar plates for application in polymer electrolyte membrane fuel cells[J]. *Materials research express*, 2017, 4(9): 095604.
- [80] Alo O A, Otunniyi I O. Electrical conductivity of polyethylene/epoxy/graphite/carbon black composites: synergy of blend immiscibility and hybrid filler[J]. *Polymer-plastics technology and engineering*, 2021, 60(13): 1-14.
- [81] Bairan A, Selamat M Z, Sahadan S N, et al. Effect of carbon nanotubes loading in multifiller polymer composite as bipolar plate for PEM fuel cell[J]. *Procedia chemistry*, 2016, 19: 91-97.
- [82] Roncaglia F, Romagnoli M, Incudini S, et al. Graphite-epoxy composites for fuel-cell bipolar plates: wet vs dry mixing and role of the design of experiment in the optimization of molding parameters[J]. *International journal of hydrogen energy*, 2021, 46(5): 4407-4416.
- [83] Glover P W J. Graphite and electrical conductivity in the lower continental crust: a review[J]. *Physics and chemistry of the earth*, 1946, 21(4): 279-287.
- [84] McKee D W. Carbon and graphite science[J]. *Annual review of materials science*, 1973, 3: 195-231.
- [85] Huttering K J. The potential of the graphite lattice[J]. *Advanced materials*, 1990, 2(8): 349-355.
- [86] Roßberg K, Trapp V. Graphite-based Bipolar Plates[M]//Handbook of Fuel Cells. Chichester: John Wiley & Sons, 2010, 1: 1-6.
- [87] Kang K, Park S, Ju H. Effects of type of graphite conductive filler on the performance of a composite bipolar plate for fuel cells[J].

- Solid state ionics*, 2014, 262; 332-336.
- [88] Heo S I, Yun J C, Oh K S, et al. Influence of particle size and shape on electrical and mechanical properties of graphite reinforced conductive polymer composites for the bipolar plate of PEM fuel cells[J]. *Advanced composite materials*, 2012, 15(1): 115-126.
- [89] Wang D, Zhang J, Li X, et al. Woven microsphere architected by carbon nanotubes as high-performance potassium ion batteries anodes[J]. *Chemical engineering journal*, 2022, 429; 132272.
- [90] Cao B, Zhang Q, Liu H, et al. Graphitic carbon nanocage as a stable and high power anode for potassium-ion batteries[J]. *Advanced energy materials*, 2018, 8(25); 1801149.
- [91] Saadat N, Dhakal H N, Jaffer S, et al. Expanded and nano-structured carbonaceous graphite for high performance anisotropic fuel cell polymer composites[J]. *Composites science and technology*, 2021, 207; 108654.
- [92] Lee M H, Kim H Y, Oh S M, et al. Structural optimization of graphite for high-performance fluorinated ethylene-propylene composites as bipolar plates[J]. *International journal of hydrogen energy*, 2018, 43(48); 21918-21927.
- [93] Yan X, Hou M, Zhang H, et al. Performance of PEMFC stack using expanded graphite bipolar plates[J]. *Journal of power sources*, 2006, 160(1); 252-257.
- [94] Fan R, Peng Y, Tian H, et al. Graphite-filled composite bipolar plates for fuel cells: material, structure, and performance[J]. *Acta physico-chimica sinica*, 2021, 37(9); 2009095.
- [95] Suherman H, Sahari J, Sulong A B. Effect of small-sized conductive filler on the properties of an epoxy composite for a bipolar plate in a PEMFC[J]. *Ceramics international*, 2013, 39(6); 7159-7166.
- [96] Bühler R, Thommen M, Le Canut J M, et al. Highly conductive polypropylene-based composites for bipolar plates for polymer electrolyte membrane fuel cells[J]. *Fuel cells*, 2021, 21(2); 155-163.
- [97] Lim J W, Lee D G. Surface modifications of gasketless carbon composite bipolar plates for gas tightness of PEM fuel cells[J]. *International journal of hydrogen energy*, 2013, 38(28); 12343-12352.
- [98] Dhakate S, Sharma S, Borah M, et al. Expanded graphite-based electrically conductive composites as bipolar plate for PEM fuel cell[J]. *International journal of hydrogen energy*, 2008, 33(23); 7146-7152.
- [99] Tang A, Crisci L, Bonville L, et al. An overview of bipolar plates in proton exchange membrane fuel cells[J]. *Journal of renewable and sustainable energy*, 2021, 13(2); 022701.
- [100] Fan R, Zheng J, Peng Y, et al. The conductive network optimization of composite graphite plates and its morphological analysis[J]. *Chemical engineering journal*, 2022, 446; 136652.