Research Progress and Prospects of Bipolar Plate Materials for Proton Exchange Membrane Fuel Cells

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Abstract. The bipolar plate is an essential component of proton exchange membrane fuel cells (PEMFC), with crucial functions such as gas separation, cooling, conduction, heat dissipation, and water ejection. The advancement of proton exchange membrane fuel cell technology relies on the performance of the bipolar plate. Recognizing the significance of bipolar plates for PEMFC functionality, researchers have been striving to create bipolar plates with high conductivity, good gas tightness, appropriate mechanical properties, corrosion resistance, and cost-effectiveness. This paper provides a detailed analysis of recent research progress on bipolar plates, aiming to contribute to the development of new and optimized products in the PEM fuel cell industry. The study in this paper focuses on three aspects: (1) Introduction to the polymer electrolyte membrane (PEM) fuel cell technology system and development directions, (2) Discussion on the structure of carbon-based fillers and their impact on the final electrical performance of composite bipolar plates, (3) Introduction to the Dual Percolation Effects of resins and Their Applications.

Keywords: PEMFC, Bipolar plates, Conductivity, Dual Percolation Effects.

1. Introduction

With the advancement of social productivity, the rapid development of the transportation industry has led to a significant consumption of fossil fuels, exacerbating energy and environmental pollution issues. The development of fuel cell technology has addressed these challenges.

Fuel cell (FC) technology applications involve the electrochemical conversion of the chemical energy of fuel (hydrogen) and oxidant (air) into electrical energy. Since the 1930s, various improvement ideas for fuel cells have been continuously proposed. Different types of fuel cells have emerged and gradually transitioned from military to civilian applications, as shown in Figure 1. Starting from the latter half of the 20th century, major automotive manufacturers have been actively researching fuel cell vehicles, and currently, a variety of high-performance fuel cell vehicle products have entered the commercialization phase worldwide. Depending on the electrolyte used, fuel cells can be categorized into various types such as Alkaline Fuel Cell (AFC), Proton Exchange Membrane Fuel Cell (PEMFC), Phosphoric Acid Fuel Cell (PAFC), Molten Carbonate Fuel Cell (MCFC), and Solid Oxide Fuel Cell (SOFC). Among these, the Proton Exchange Membrane Fuel Cell (PEMFC) is the most popular efficient alternative energy source due to its high power density, efficiency, low operating temperature, ease of fuel supply, low noise, quick start-up time, and long lifespan.

Proton Exchange Membrane Fuel Cells are sometimes also referred to as Polymer Electrolyte Membrane Fuel Cells (PEMFC). The structure of PEMFC mainly consists of two bipolar plates and a Membrane Electrode Assembly (MEA), which collectively establish the flow fields for hydrogen, oxygen, and coolant in the system, as shown in Figure 2 [1] [2].

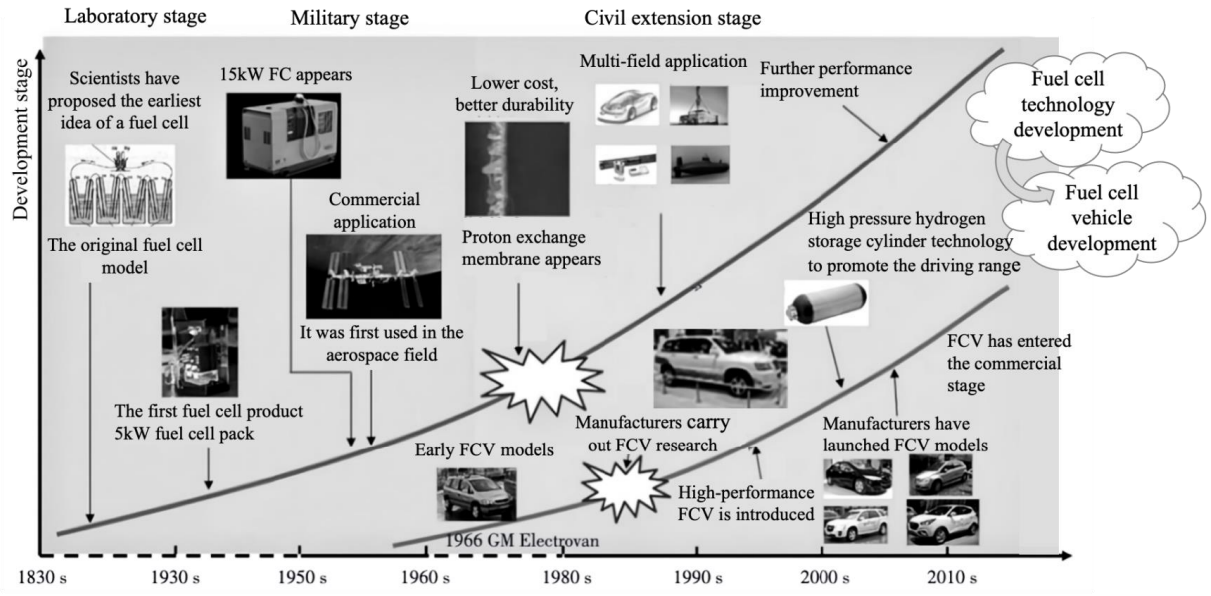

Figure 1. Development Chart of Fuel Cell Technology

PEMFC mainly consists of the stack and system components (such as the air compressor, humidifier, hydrogen circulation pump, and hydrogen tank); the stack system functions as the core operation, comprising individual cells made up of membrane electrodes, bipolar plates, as well as current collectors, end plates, sealing rings, and other structures. In recent years, research in hydrogen fuel cell technology has primarily focused on the stack, bipolar plates, and control technologies, as illustrated in Figure 1.

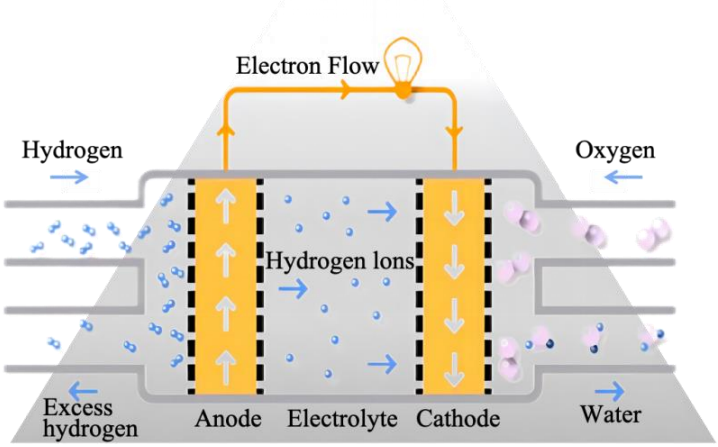

Figure 2. Schematic Diagram of Proton Exchange Membrane Fuel Cell Structure

While the development and promotion of Proton Exchange Membrane Fuel Cells can significantly reduce the use of traditional fossil fuels and environmental pollution, the commercialization path of PEMFC fuel cells remains challenging, particularly in terms of durability, reliability, and cost. Therefore, there is an urgent need to reduce the material costs of fuel cell manufacturing to enable broader commercial applications. One of the fundamental components of PEMFC is the bipolar plate (BP). BPs play a crucial role in the equipment, accounting for approximately 60-80% of the total cell weight and 40-50% of the cost.[3] Hence, cost reduction strategies related to bipolar plates hold significant promise for enhancing commercial viability. BPs serve multiple functions, including connecting each cell, supplying reactant gases to the anode and cathode, and removing reaction products from the cell. As current collectors, BPs must possess high conductivity and robust mechanical properties to withstand clamping forces. Depending on the type of matrix material used, BPs can be categorized as graphite BPs, metal BPs, and composite BPs, as shown in Figure 3.

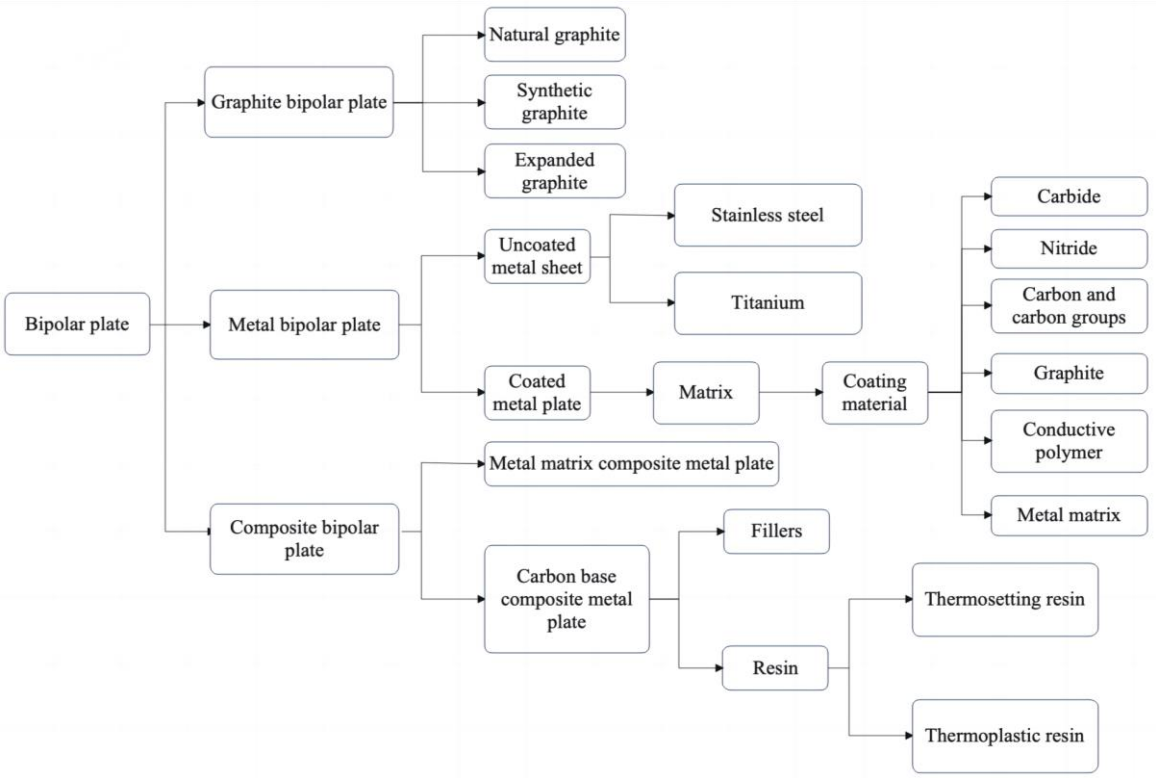

Figure 3. Types of bipolar plates

Each type of bipolar plate has its own advantages, but also has noticeable drawbacks, as shown in Table 1.

Table 1. Advantages and Disadvantages of different bipolar plates materials

Type	Materials	Advantages	Disadvantages
Carbon	Graphite bipolar plates	Low density, good corrosion resistance, high strength	High porosity, low mechanical strength, high brittleness, difficult for mass production, high price
	Composite bipolar plates	Excellent corrosion resistance, low density	Poor electrical conductivity, high porosity, high price
Metal	Metallic bipolar plates	bending strength, low gas permeability, better workability, low cost	Heavy, susceptible to surface corrosion

More specifically, traditionally, materials such as metals and graphite have been renowned for their excellent conductivity and thermal properties and have been used in the manufacturing of bipolar plates. Graphite BPs exhibit outstanding conductivity and corrosion resistance, with the technology being the most mature and graphite being the most widely used carbon material for BP commercial applications. However, their brittleness leads to insufficient mechanical strength, porous surface structures that may result in the mutual permeation of hydrogen and oxygen. Correcting these issues by increasing the thickness of graphite bipolar plates makes it challenging to reduce the thickness, directly impacting the cost of the cell and making their application difficult in compact and impact-resistant scenarios. Therefore, metal BPs with superior performance and cost advantages have become a focal point of development. Mainstream metal BPs have a thickness not exceeding 0.2mm, significantly reducing volume and mass, markedly increasing stack power density, and possessing good ductility, excellent conductivity, thermal properties, and high fracture toughness. Currently, major hydrogen fuel cell vehicle companies (such as Honda, Toyota, General Motors, among others) employ metal BP products. However, metal BPs exhibit poor corrosion resistance, with metals easily dissolving in acidic environments, potentially poisoning the membrane electrode assembly with

leached ions. As metal ion solubility increases, the ohmic resistance rises, reducing the cell's output power and adversely affecting the overall performance of PEM fuel cells.

Consequently, recent research efforts have been focused on developing cost-effective and viable alternative materials. Numerous studies have explored polymer composite bipolar plates, investigating a range of formulations containing thermosetting and thermoplastic resins combined with various carbon fillers, including graphite, carbon black, carbon fibers, and carbon nanotubes. Due to the scarcity of review papers introducing the latest materials, rigorous scrutiny and summarization of research in these areas are necessary.

To promote the commercialization of PEMFC, the Fuel Cell Technologies Office of Energy, Department of Energy (DOE), has proposed bipolar plate technology parameter goals for 2025, as shown in Table 2.[4] [5]

Table 2. 2025 US Department of Energy (DOE) standard for bipolar plates

Characteristic	Units	2025 Targets
Cost	$/kW	2
Plate weight	kg/kW	0.18
Plate H2 permeation	Std $cm^3/(sec\ cm^2\ Pa)$@ 80°C	2×10^{-6}
coefficient	3 atm,100% RH	
Corrosion, anode	$\mu A/cm^2$	less than 1 and no active peak
Corrosion, cathode	$\mu A/cm^2$	less than 1
Areal specific	Ohm cm^2	less than 0.01
resistance		
Flexural strength	MPa	> 40

2. Materials

Over the past few decades, extensive research has been conducted to enhance performance and reduce costs by minimizing volume and weight. Composite Bipolar Plates (CBP) have garnered attention due to their low manufacturing costs and excellent corrosion resistance. Typically, CBPs are composed of insulating resins and conductive fillers, with the conductive fillers bonded by insulating resins. However, due to the conflicting demands between conductivity and mechanical properties, their use in BPs is less than other materials. [6] [7] In the last decade, efforts have been made to enhance mechanical strength and conductivity from the perspectives of material design and manufacturing process technology application. In terms of materials, fillers contribute more significantly to electrical conductivity and other electrochemical properties, including natural graphite (NG), [8] carbon black (CB), [9] graphene, [10] carbon nanotubes (CNT), [11] and conjugated polymers such as poly (1, 4-phenylene sulfide) and polypyrrole. [12-15] Fillers have shifted from traditional carbon/epoxy composites to carbon fibers with conductive characteristics, enhancing battery performance, improving conductivity stability, and reducing production costs. On the other hand, resin matrices contribute more to mechanical properties and costs. Commonly used resins include epoxy resins, phenolic resins, vinyl esters (VE), polypropylene (PP), polyethylene (PE), polyphenylene sulfide (PPS), among others [16-20]. During processing, surface modification methods for bipolar plates can enhance their interaction with electrodes. Jiang [21] et al. improved the electrical performance of bipolar plates using cactus-like carbon nanofibers, while Jeong [22] et al. enhanced interface contact resistance through electrode integration with bipolar plates. Traditional carbon/epoxy composites form resin-rich regions on the surface. Among surface treatment methods, the soft layer technique is considered the most suitable for commercialization, significantly reducing ASR data without the need for coatings or post-processing techniques.

In summary, fillers play a greater role in electrical conductivity and other electrochemical properties, while resin matrices contribute more to mechanical properties and costs. The toughening effect of fillers can easily compensate for the toughness loss of brittle epoxy resins based on graphite

matrices, while enhancing gas tightness/mechanical strength through resin reinforcement. The long-term stability of chemical, thermal, mechanical, and bipolar plates is a key criterion for fuel cells to maintain long-term operation. Therefore, greater attention should be paid to feasible alternative materials that can promote strength, corrosion resistance, ease of mass production, low porosity, and cost-effectiveness. This section will further explore different types of material selection and their potential advantages in bipolar plate applications. Thus, this section discusses the progress in electrical conductivity research of CBP materials from the perspectives of carbon-based materials and other materials.

2.1. Fillers

In PEMFCs, the primary function of CBP is to collect and transfer electrons between components, reducing ohmic polarization losses requires enhancing the high conductivity of CBP. It is crucial to improve the conductivity of CBP by forming a continuous conductive network through conductive fillers. Conductive fillers typically refer to particles or fibers added to the base material to enhance the material's properties or performance. Commonly used conductive fillers include graphite (Gr), carbon fibers (CF), carbon black (CB), and carbon nanotubes (CNT).

2.1.1. Graphite(Gr)

Graphite, with its low density and affordable price, is a commonly used material for bipolar plates. The graphite lattice consists of multiple layers of two-dimensional graphene sheets tightly bonded with sp2 hybridized carbon atoms forming hexagonal rings, as shown in Figure 4. Each carbon atom in the lattice has three neighboring carbon atoms at the vertices of an equilateral triangle. If the interlayer 2pz orbitals are parallel, they overlap. The graphite lattice is formed by stacking parallel two-dimensional (2D) thin sheets, with hybridized carbon atoms tightly bonded in hexagonal rings, and π orbitals distributed throughout the graphite sheet, imparting thermal and electrical conductivity. Adjacent graphite sheets are held together by weak van der Waals forces, allowing them to easily slide past each other, leading to the brittleness of graphite.[23-24]

In large planar layers, there are intermediate bonds between atoms and metal bonds, leading to excellent conductivity and good thermal conductivity, which is crucial for the transfer and dispersion of heat generated in batteries. In addition to its metallic properties like conductivity and high thermal conductivity, graphite also possesses high corrosion resistance and inert non-metallic properties. Generally, graphite is categorized based on its formation process into natural graphite (NG), synthetic graphite (SG), and expanded graphite (EG). Sengupta et al. reviewed the performance of graphite and synthetic graphite in carbon polymer composite materials.[25-27]

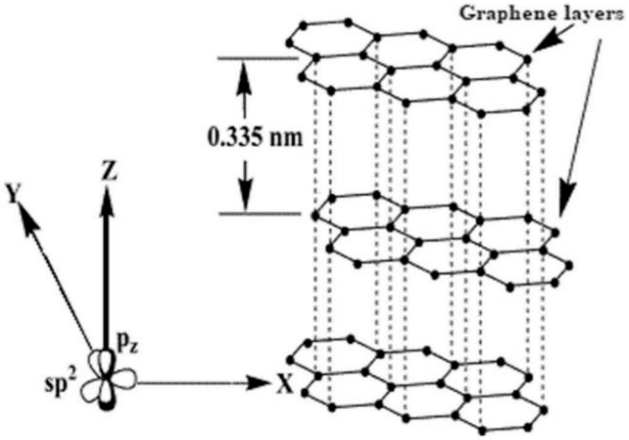

Figure 4. Graphite structure showing sp^2 hybridized atoms and layers [27]

In filling high-insulation polymer matrix materials with conductive polymers, achieving uniform dispersion of the conductive polymer within the matrix is essential. This dispersion allows the conductive macromolecules to fully extend throughout the matrix, forming complete conductive pathways, significantly enhancing conductivity. The size and shape of the conductive particles also

impact the electrical properties of the final composite material. Table 3 describes how different sizes and shapes of graphite affect various properties of bipolar plate composite materials. Studies indicate that the size of graphite influences the performance of bipolar plates; as graphite size decreases, the bending strength of bipolar plates increases while the electrical conductivity gradually decreases. Due to the dimensional differences between flake graphite and spherical graphite, two-dimensional flake graphite is more likely to form a conductive network in space compared to spherical graphite, resulting in higher electrical conductivity in flake graphite composites than in spherical composites. However, spherical composites have a higher filling capacity, improving processability by at least 5 to 12 times. Generally, the conductivity threshold of spherical particles is lower than that of fibrous particles. Moreover, a higher aspect ratio of particles leads to the formation of more effective conductive networks. Essentially, a higher aspect ratio in particle structure favors the formation of efficient conductive pathways. [28-31]

Table 3. Comparison between graphite materials with different sizes and shapes and their influence on different attributes of the BPP composite

Graphite Particle Size (μm)	Particle Shape	Resin-Process	Max. Conductivity (S/ cm)	Other Attributes (Max. value)	Determinant Factors	Ref.
<53–>177	–	Vinyl ester-Bulk molding compound	153.8	Porosity,0.57%, Flexural strength, 31.23 MPa,Oxygen permeability 6.76 × 10^8 cm^3 / cm^2s	Comparison between conductivity, flexural strength, corrosion current and permeability for different sizes. The bigger particles yield better properties	[28]
5,10&20	Flake like and Spherical	Polypropylene-Injection and compression molding	96.9 (Through-plane)	Output rate, 12 kg/h(during compounding)	Comparison between shapes and processability with output rate. Optimization required between the conductivity and process output	[29]
10,20&40	Flake and lumps	Phenolic resin-Compression molding	146	Flexural strength, 44 MPa	Comparison between shapes and types (SG and NG) Bigger flake type NG showed optimum properties due to higher aspect ratio and crystallinity	[30]
7,15&25	Sphere and flake type	Phenol powder-Compression molding	~250	Flexural strength, >50 MPa	Comparison between size, shape and molding pressure.Flake-type found to be superior for composite properties	[31]

Another important aspect of graphite is its conductivity along its crystal layer structure, but perpendicular to the plane, its conductivity is very low, affecting the conductivity of composite materials in the plane direction. Kim [32] et al. studied the synergistic effects of hybrid carbon systems of graphite powder and continuous carbon fiber fabrics and observed optimal conductivity at filler contents of 70-75 vol%.

Currently, the influence of graphite particle size and graphite ratio on CBP performance has been extensively researched. For example, Rafi et al. have demonstrated that larger NG particle sizes can enhance CBP density, electrical conductivity, and flexural strength. When the particle size increases from 45 μm to 90 μm, the electrical conductivity of CBP increases from around 40 S/cm to 100 S/cm.

This phenomenon can be attributed to the reduced contact resistance of graphite when using larger particle sizes. [33-34]

Expanded graphite (EG) is a modified natural graphite with high conductivity, having a lower density (0.01-0.015 g/cm³) than natural graphite (2.2 g/cm³). Therefore, it can be considered as one of the alternative materials for developing composite bipolar plates. Compared to traditional graphite and synthetic graphite, expanded graphite exhibits excellent formability and conductivity due to its larger interlayer spacing, making it lighter with a higher aspect ratio, reducing interlayer connections and forming a more efficient electrical network. A.R. Dhakate prepared carbon-filled epoxy composites using phenolic resin and EG. Chemical intercalation of EG was performed on natural graphite of 270 μm in size, resulting in a CBP volume density of 1.5 g/cm³ with electrical and mechanical properties reaching 120 S/cm and 54 MPa, respectively. The excellent performance of CBP at high resin content is attributed to the porous structure of EG, facilitating resin penetration and the formation of a conductive network.[35-36]

However, there are some issues such as insufficient plate strength and poor gas isolation. [37][38] It is necessary to add additional polymer resin during the pressing process to enhance plate strength and improve gas tightness. This poses a dilemma in the composition of expanded graphite bipolar plate materials as high conductivity requires more mixed expanded graphite components while high gas tightness and strength require a higher resin content. To address this issue, Liang Zhao[39] et al. developed an EG-based conductive composite bipolar plate suitable for PEM fuel cells by adjusting the ratio of EG to resin. Introducing support materials can effectively improve the strength of graphite bipolar plates. Bi [60]et al. compacted multiple flexible graphite foils supported by polycarbonate, resulting in a significantly improved support for the cell stack. Ji [40]et al. combined graphite sheets with support material boards to enhance the strength of the bipolar plates, increasing current density and cell voltage. To address the issue of poor gas tightness, impregnating graphite plates is an effective method. Wang [41]et al. impregnated graphite bipolar plates with a concentrated water glass solution using a vacuum compression method, converting it to SiO2, significantly reducing the porosity. However, during the forming process, cracks may appear due to changes in resin hardness. Therefore, the selection of resin type must be tailored to the specific circumstances.

In polymer composites reinforced with expanded graphite (EG), high aspect ratio conductive particles also have a beneficial impact on reducing the permeation threshold of CPCs. EG is typically produced by first treating natural graphite with acid to generate graphite intercalation compounds. This compound is then rapidly heated to expand the interlayer structure. EG retains the layered structure of natural graphite flakes but possesses pores of various sizes and nano flakes with very high aspect ratios. The porous network favors physical and chemical adsorption between EG and polymers, while the high aspect ratio benefits electrical contact between adjacent EG particles. Zheng and Wong[42] studied the electrical behavior of PMMA-based composites reinforced with carbon black, EG, and unmodified graphite particles. They observed that high aspect ratio EG particles provided a penetration threshold of 1 wt.%, while for carbon black and unmodified graphite, these values were 8.0 wt.% and 3.5 wt.%, respectively. Zhao [43]et al. also reported a penetration threshold of only 1% for PPS-EG composites.

In conclusion, researchers have addressed various drawbacks of graphite bipolar plates through different means. However, graphite bipolar plates are typically thick, leading to larger volume and weight, which hinders the commercialization of PEMFCs. These issues limit the application of graphite bipolar plates in PEMFCs.

2.1.2. Carbon fiber (CF)

Carbon fiber is a synthetic material with a carbon content exceeding 90%, renowned for its low density, good conductivity, excellent mechanical properties, and chemical stability.[44] Currently, its primary precursor for production is polyacrylonitrile (PAN). It possesses significant dimensions and a high aspect ratio, contributing to its exceptional strength and stiffness. The microcrystalline structure of carbon fiber gives it high strength and modulus along the fiber axis, making it suitable as an auxiliary conductive filler to enhance the mechanical strength of bipolar plates. [45-46]

Maheshwari [47] utilized carbon fiber-reinforced phenolic resin/graphite composite materials for bipolar plate fabrication. The results showed that carbon fiber effectively enhanced the flexural and compressive strength, with a bending strength of 82 MPa observed when the carbon fiber content was 10 wt.%, representing a 140% improvement over pure phenolic resin graphite composite materials.

Researchers also employ carbon fiber to enhance electrical performance. Lee [48] et al. studied the performance of polymer composite bipolar plates prepared using nonwoven carbon fiber felts, demonstrating that carbon fiber felts not only imparted high mechanical performance to the bipolar plates but also provided them with high electrical conductivity. Similar research by Jiang [49] et al. indicated that carbon fiber fabrics could enhance the electrical and mechanical properties of composite bipolar plates. From the results of these studies, it is evident that using carbon fiber or carbon felts as conductive fillers or additives can improve the performance of composite material bipolar plates, as carbon fiber is a high aspect ratio filler that easily forms a good conductive network. [50-51] The increased electrical conductivity depends on the aspect ratio, with higher aspect ratios leading to lower permeation thresholds in composite materials. However, increasing the aspect ratio has some drawbacks, such as increased porosity affecting the hydrogen permeability of the bipolar plates, as illustrated in Figure 5. Additionally, the aggregation of carbon fibers increases the volume resistivity of the composite material, necessitating the optimization of carbon fiber content. Nevertheless, during the fabrication of these composite material bipolar plates, the poor flowability of raw materials containing carbon fibers when heated to a molten state and the complex carbon felt laying process during plate formation make controlling the final manufacturing process challenging.

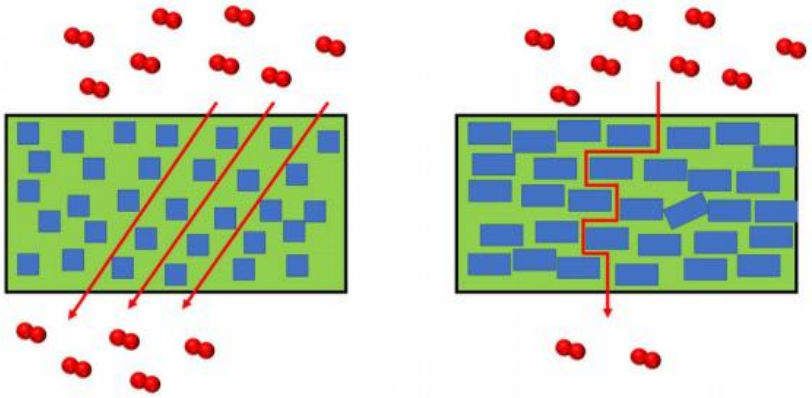

Figure 5. Effect of the aspect ratio of filler on the alr tightness of composite bipolar plates. [60]

2.1.3. Carbon black(CB)

Carbon black (CB) is a synthetic elemental carbon primarily produced through electric furnace or acetylene processes. [47, 52] This material offers advantages such as durable and stable conductivity, affordability, and abundant resources. [53] Although it has lower mechanical properties and conductivity, the addition of carbon black can effectively enhance the material's conductivity. With an increase in the amount of carbon black, the inter-particle spacing decreases, leading to the formation of numerous conductive network pathways when the material is close or in contact, subsequently reducing the material's resistance and improving its conductivity. Among various fillers, carbon black has the most significant impact on the conductivity of bipolar plates. Generally, as the volume fraction of carbon black increases, the permeability of the composite bipolar plate decreases while the conductivity increases. Kim [54] et al. innovatively dispersed carbon black uniformly onto carbon fabric/phenolic resin composite bipolar plates using an impregnation method. Furthermore, they subjected the bipolar plate surfaces to propane flame treatment to reduce interface contact resistance. Studies have shown that incorporating 4 wt% of carbon black into the composite bipolar plates, followed by a 5-second propane flame treatment, significantly reduced the interface contact resistance to 27 $m\Omega cm^2$, exhibiting excellent electrical performance (Figure 6(a) and 6(b)). However, it is worth noting that propane flame treatment of the composite bipolar plates leads to increased

permeability due to volume shrinkage caused by surface carbonization and the formation of cracks and voids within the matrix.

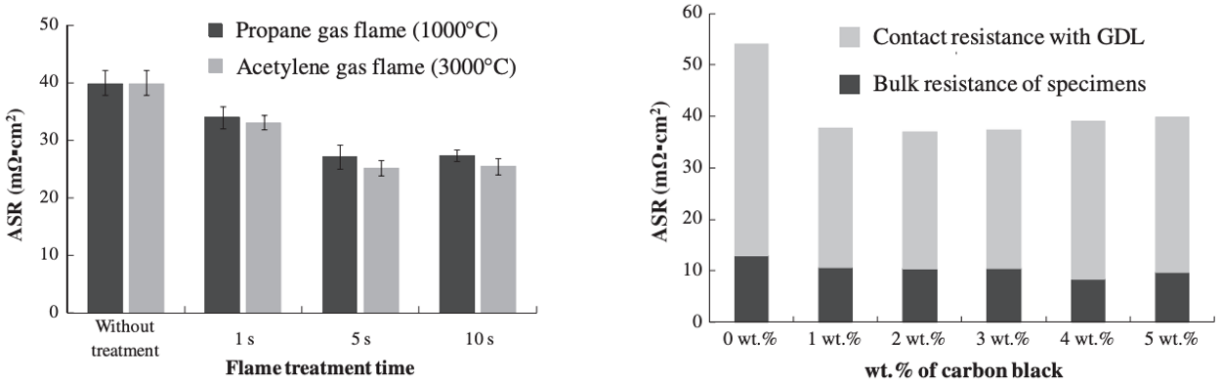

Figure 6. (a) ASR of the carbon fabric/phenolic composite bipolar plates with respect to the wt.%of carbon black under the compaction pressure of 1.5 MPa. (b) ASR of the flame-treated carbon fabric/phenolic composite bipolar plates with respect to flame treatment time under the compaction pressure of 1.5 Mpa [54].

Gautam [55] et al. used phenol formaldehyde (PF) as a binder, expanded graphite (EG) as a conductive matrix, and carbon black and graphite powder as conductive additives to prepare composite bipolar plates. The research findings indicated that PF/EG/carbon black/graphite powder composite bipolar plates exhibited better conductivity and mechanical properties compared to bipolar plates without carbon black. When the EG content was 35%, carbon black addition was 5%, and graphite powder addition was 3%, the composite bipolar plates achieved a high in-plane electrical conductivity of 374.42 S/cm and a bending strength of 61.82 MPa, with superior single-cell performance compared to composite bipolar plates without carbon black.

However, when the volume fraction of carbon black exceeded a critical level, the conductivity of the composite bipolar plates decreased. This could be attributed to poor wetting between carbon black and other particles leading to particle aggregation, as shown in Figure 7. Lee [56] et al. and Dhakate [57] et al. confirmed this observation. They investigated the effect of carbon black mass fraction on the electrical conductivity of composite bipolar plates using carbon black as a conductive filler. Lee [56] et al.'s research indicated that the composite bipolar plates exhibited the highest electrical conductivity of 158 S/cm when the volume fraction of carbon black was 5 vol%. Dhakate [57] et al.'s results showed a 45% increase in conductivity when the carbon black loading reached 5 wt%. The increase in conductivity was attributed to the formation of additional conductive pathways between small carbon black particles and large EG particles. Beyond the critical concentration, as the volume fraction of carbon black continued to increase, carbon black aggregation led to a reduction in electron migration pathways, an increase in resistivity, and a decrease in conductivity. Suherman [58] et al. suggested that a low resin content insufficient to encapsulate carbon black and graphite matrices could also affect the conductivity and mechanical properties of composite bipolar plates.

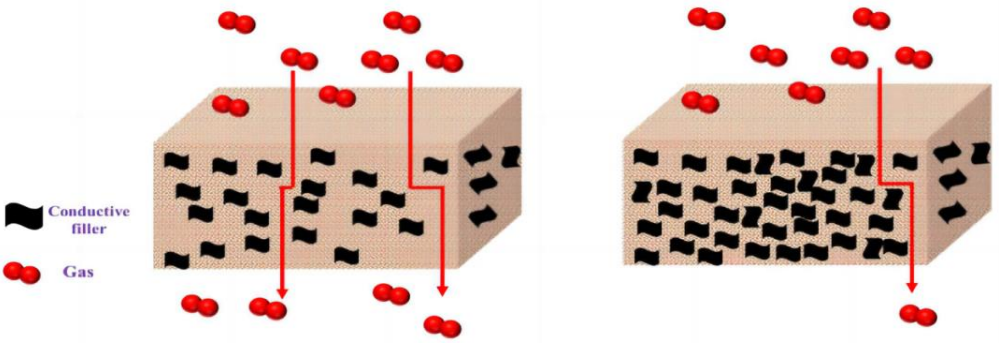

Figure 7. Effect of conductive filler concentration on air tightness of composite bipolar plates.

Furthermore, a compromise between electrical and mechanical performance should be considered. When the conductivity reaches a threshold, excessive filler cannot enhance the conductivity of composite bipolar plates (CBP); instead, it can lead to a decrease in the mechanical properties of CBP. Unlike the addition of carbon black (CB) as a secondary filler, when the CB content increases by 2 wt.%, the composite material's bending strength increases. However, with further additions of CB content, the bending strength of the composite material continues to decrease, as shown in Figure 8. The maximum bending strength obtained when the mass fraction of carbon black is 2% is 75.6 MPa. Initially, the formation of voids occurs as the gaps between carbon fiber (CF) and carbon black reduce the bonding between the filler and the matrix, resulting in poor mechanical strength, as reported in other research results as well. The trend in the change of bending strength of the composite material is similar to the trend in the change of electrical conductivity after the addition of carbon black. [59-63]

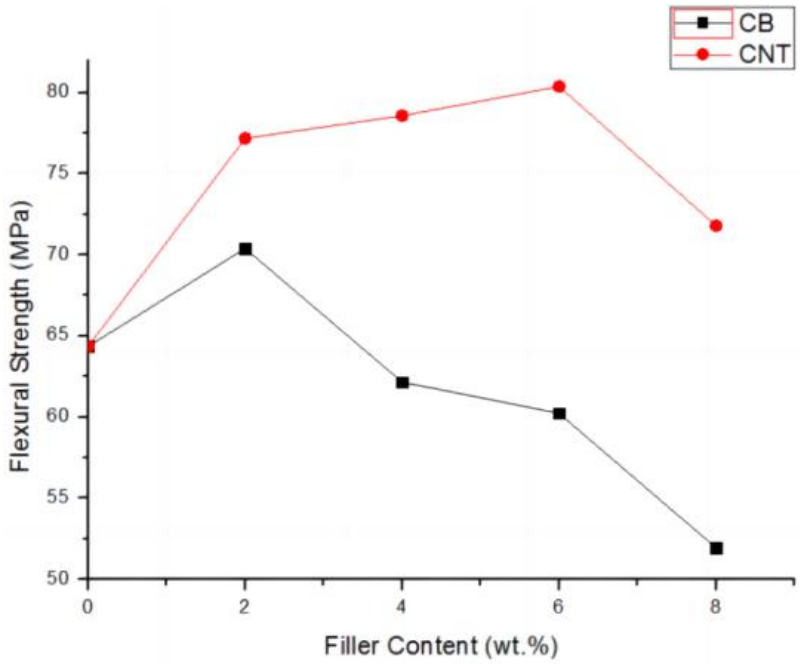

Figure 8. Effect of adding secondary filler on flexural strength [59]

Carbon black, as an effective conductive material, has found wide applications in composite bipolar plates due to its low cost, small particle size, and significantly lower resistivity compared to the bipolar plates themselves. Researchers have also investigated the influence of carbon black loading on the electrical performance of composite bipolar plates, providing valuable insights for future studies. However, there is currently a lack of in-depth exploration regarding the relationship between carbon black particle size and the electrical properties of composite bipolar plates. Therefore, this remains an area that can be further explored in future research.

2.1.4. Carbon nanotubes(CNT)

Carbon nanotubes are nano-sized particles with a tubular structure, classified into single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs). They typically range in length from 1 micron to several centimeters and in diameter from 1 to 50 nanometers. Carbon nanotubes exhibit high elastic modulus and a high aspect ratio. [64] The high elastic modulus enhances the mechanical strength of materials, while the significant aspect ratio facilitates effective electron transfer, promoting the formation of a conductive network. Therefore, carbon nanotubes have garnered considerable interest as excellent auxiliary conductive fillers in the scientific community.

The overlapping π orbitals of CNTs enable rapid electron delocalization, enhancing the conductivity of composite materials.[65] Tariq and colleagues [66] reported a 127% increase in conductivity (49.25 S/cm) by adding 2 wt% MWCNTs to PP-graphite bipolar plates. [67] The

conductivity enhancement is attributed to the uniformity of the filler, high aspect ratio, and elongated structure of MWCNTs, facilitating effective interconnection of nanoparticles and reducing resistance.

Selamat [68] et al. produced bpp using 20 wt% PP, 25 wt% CB, and varying amounts of MWCNTs and GR (ranging from 0 wt% to 0.8 wt%). Initially, the addition of 0.2 wt% MWCNTs improved the conductivity, but beyond this point, due to the clustering of high concentrations of MWCNTs, the conductivity continued to decrease. The conductivity was 179.42 S/cm without added MWCNTs, 285.78 S/cm with 0.2 wt% MWCNTs, and 42.09 S/cm with 0.8 wt% MWCNTs.

Radzuan [69] et al. developed nanocomposite materials composed of PP/CF/MWCNTs using a compression molding method and investigated the effect of CF content on electrical performance. They varied the CF content from 65 wt% to 70 wt% while maintaining MWCNTs at 5 wt%. At a CF content of 70%, the maximum electrical conductivity of the composite material reached 11.128 S/cm.

In another study by Bouatia [70] et al., PET/GR/CB nanocomposite materials were prepared, with some samples incorporating 5 wt% MWCNTs to explore synergistic effects. Equivalent amounts of MWCNTs were used to replace 5 wt% of carbon black and GR to reduce surface resistivity. However, when the total concentration of carbon black, GR, and MWCNTs approached 30 wt%, the decrease in resistivity became less significant. This suggests that lower concentrations of MWCNTs promote faster connections between CB and GR particles compared to higher concentrations.

S.R. Dhakate [71] et al. added different volume percentages of multi-walled carbon nanotubes (MWNT) to graphite-polymer composite bipolar plates. They found that adding 1 vol% MWNT to graphite composite plates increased the electrical conductivity and thermal conductivity of the nanocomposite material by 100%. With 1 vol% MWNT, the thermal conductivity of the nanocomposite plate increased from 1 W/m K to 13 W/m K, and the in-plane thermal conductivity increased from 25 W/m K to 50 W/m K. This significant enhancement was attributed to the orientation of multi-walled carbon nanotubes in all directions in the composite material, positive synergistic effects of multi-walled carbon nanotubes, and heat transfer along the axial direction.

The addition of carbon nanotubes also helps improve the mechanical properties of CBP. Recent studies have shown that adding carbon nanotubes to single-filler graphite/EP composite materials at a filling level of 5 wt% can achieve a bending strength of 46 MPa. Darıcık [72] et al. filled MWCNT particles into carbon fiber epoxy composite laminate at weights ranging from 0.1 to 2.5% and conducted electrical, mechanical, and performance characteristic tests. In the composite material sample modified with 1.25% carbon nanotubes, the in-plane electrical conductivity reached 120.05 S/cm. Considering the importance of bending strength in bipolar plates, the addition of CNTs is beneficial for the durability of bipolar plates. Therefore, the good dispersion of carbon nanotubes between CP/EP composite materials helps enhance the mechanical properties of the composite material. [73]

However, carbon nanotubes also have drawbacks, including high cost and challenges in dispersing them within a polymer matrix due to their high surface energy and nanoscale diameter. Compared to single-filler composite materials, carbon nanotubes can significantly improve the bending strength and electrical conductivity (in-plane and out-of-plane) of composite materials when effectively dispersed. There exists a threshold concentration. For instance, Suherman et al. concluded that the critical weight percentage of carbon nanotubes is 5 wt% as a secondary filler in a 20 wt% epoxy resin matrix (Figure 9). Beyond this threshold, both conductivity and strength notably decrease. Based on structural differences and cost-effectiveness of carbon nanotubes, further research is needed to explore the use of different types of carbon nanotubes in BPP. [74-77]

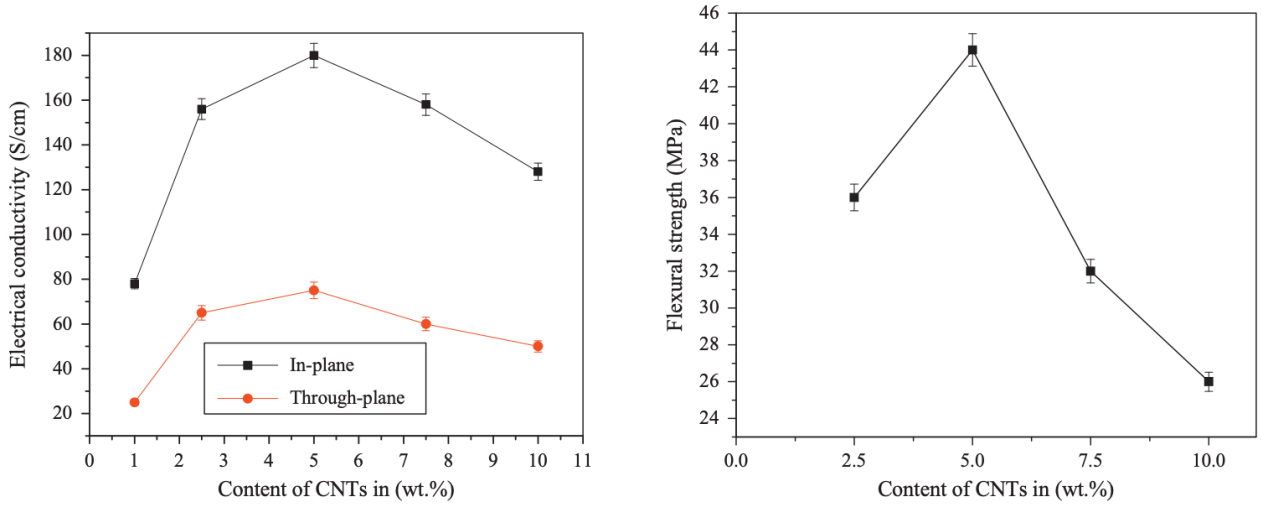

Figure 9. Effects of CNT content on a)electrical and b)mechanical properties of the BPP composite [77]

2.2. Polymers

The mechanical performance is crucial for bipolar plates (CBP) as it determines their suitability for use in PEMFCs. [78] This section provides detailed insights into widely used polymers such as thermosets and thermoplastics, along with their surface modifications, to guide in enhancing the material's mechanical properties.

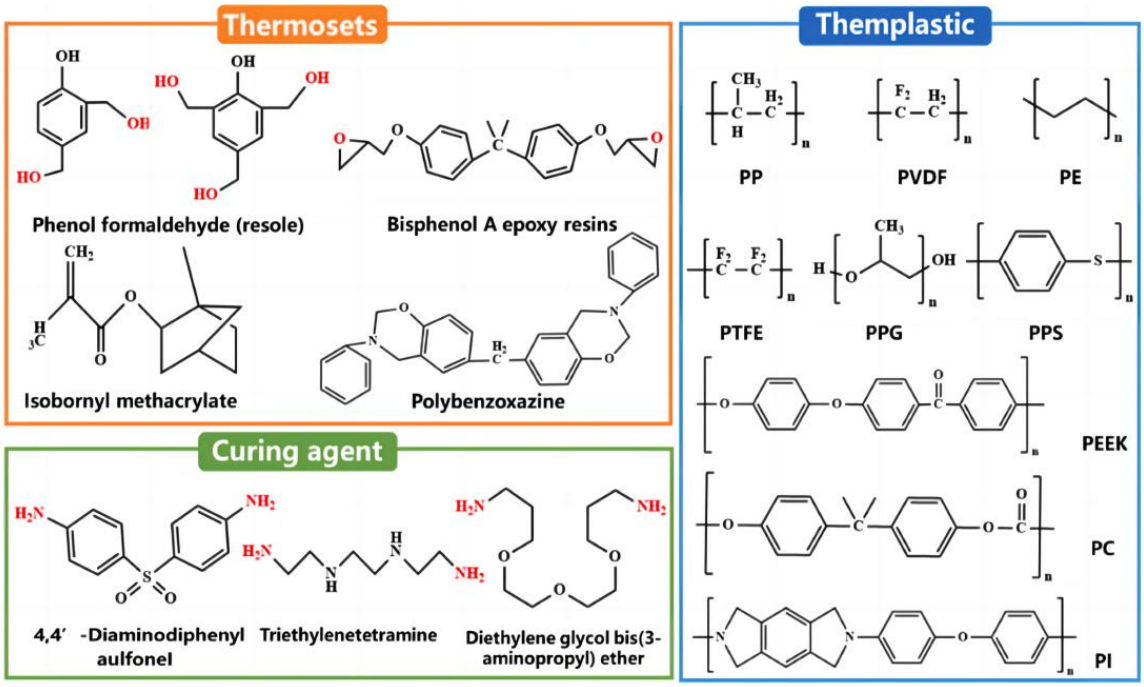

Figure 10. The representative thermosets,curing agent and thermoplastics.

Polymer resins play a vital role in the preparation of composite bipolar plates, primarily categorized into two major classes: thermosetting resins and thermoplastic resins. Common examples of primary polymer matrices include epoxy resins, vinyl ester resins, and phenolic resins for thermosetting resins, and the thermoplastic polymer family including polyvinylidene fluoride (PVDF), polypropylene (PP), polyethylene (PE), and polyphenylene sulfide (PPS). Thermosetting resins are widely used in this field due to their higher hardness, excellent stability under high-temperature fuel cell operating conditions, and lower viscosity compared to thermoplastic materials. The molecular structures of commonly used thermoplastic and thermosetting resins in CBP are illustrated in Figure 10.

The mechanical/thermal properties of thermosetting polymers are stable due to the strong interactions between polymer chains. The three-dimensional network structure of resins is formed by chemical reactions between small molecules. With small resin molecules, low viscosity, good flowability, and ease of uniform dispersion between conductive fillers, these advantages are highly suitable for manufacturing CBP. Phenolic resins and epoxy resins, as the most important thermosetting materials, have found wide applications in CBP due to their excellent thermal and mechanical properties, chemical resistance, dimensional stability, and processability. For instance, A. Hosh prepared CBP using a solution intercalation method by thoroughly mixing a certain amount of carbon-based material (75%) with a phenolic resin binder mechanically, followed by CBP preparation using a hot pressing method. The results indicated that as the molding temperature increased from 170°C to 200°C, the mechanical performance and electrical conductivity of the alloy decreased from 60 MPa to 10 MPa and from 425 S/cm to 300 S/cm, respectively, attributed to the changes in the resin network structure at high temperatures. [79-84]

This phenomenon can be attributed to the thermal degradation and volatilization of diphenyl ether in the resin structure, leading to a significant decrease in the mechanical and electrical properties of the composite material. Minkook Kim proposed a flame treatment method for CBP and subjected CBP with phenolic resin as a binder to treatment at different temperatures. Experimental results showed that the contact resistance of CBP decreased with the carbonization of the polymer, attributed to the exposure of conductive fillers during the carbonization process. However, as the polymer carbonizes, the bending strength of CBP inevitably decreases. [85-87]

At room temperature, thermoplastic plastics are high-molecular-weight solids that are linear or have minimal branching.

There are no cross-links between molecules in thermoplastic resins; they are only attracted to each other through van der Waals forces or hydrogen bonds. The substructure of thermoplastic resins is linear, exhibiting characteristics of thermal softening and cold hardening, with no chemical reactions occurring during hardening. Therefore, thermoplastic materials disperse in fillers through melt blending. Thermoplastic resins are more widely used in CBP compared to thermosetting resins. Common thermoplastic materials used in CBP include polypropylene (PP), polyethylene (PE), polyphenylene sulfide (PPS), polycarbonate (PC), and polyvinylidene fluoride (PVDF). For instance, N. Stabler considered compounds of polyvinylidene fluoride (PVDF), polyphenylene sulfide (PPS), and polypropylene glycol (PPG), densely mixing high contents of graphite (85%) with polymers in a compounding machine, then molding these mixtures into CBP using high-temperature molding. CBP with PPS as a binder exhibits high bending strength of 35 MPa, with minimal impact on its performance from methanol. CBP with PPS as a binder shows excellent mechanical properties, mainly attributed to the π-π stacking interactions between graphite and aromatic rings in PPS, improving the contact between filler and binder compared to other saturated resin network structures. The electrical conductivity of PPG composite materials is less than 50 S/cm, with the significant difference in conductivity attributed to the numerous hydroxyl groups in the resin structure leading to resin aggregation, resulting in uneven distribution and lower electrical conductivity of PPG. [88-94]

The application of thermoplastic resins is limited by their glass transition temperature (Tg). For example, although polypropylene exhibits excellent corrosion resistance and impact resistance, its lower Tg makes it unsuitable for manufacturing high-temperature fuel cell bipolar plates. In the preparation of composite bipolar plates, the mixture of resin and conductive fillers can enhance the material's flexural strength and gas permeability. Thermoplastic resins, when mixed with conductive fillers, are suitable for compression molding but require longer cooling times. In contrast, thermosetting resins can be compression molded without the need for cooling, simplifying the manufacturing process and shortening the production cycle.

Additionally, thermoplastic resins offer high processing repeatability and molding efficiency. However, composite materials made with them tend to be relatively brittle, often requiring greater thickness to ensure sufficient mechanical strength. In contrast, thermosetting resins can form a strong

three-dimensional network structure, providing higher bending strength and allowing for the manufacture of thinner bipolar plates. Kakati [95] et al. mixed graphite, carbon black, and carbon fibers with two types of phenolic resins and used molding techniques to produce composite bipolar plates. The study found that thermoplastic phenolic resins and thermosetting phenolic resins can react with the curing agent hexamethylenetetramine at high temperatures, forming large polymer molecules with a three-dimensional network structure, significantly enhancing the performance of the bipolar plate material. Experimental results have shown that bipolar plates made of thermoplastic resins have higher conductivity but lower strength and higher porosity. Therefore, considering the application environment of bipolar plates, composite plates made of thermosetting phenolic resins are more suitable for fuel cell bipolar plates.

3. Synergistic Effects of Conductive fillers

In this chapter, we will explore the synergistic effects of different types of conductive additives on the performance of composite materials. A common practice aimed at enhancing the performance of composite materials is the addition of secondary fillers alongside graphite. The synergistic interactions between materials play a crucial role in improving the performance of bipolar plates. By optimizing the material composition and the form of synergy in bipolar plates, conductivity, mechanical strength, and thermal stability can be enhanced.

3.1. The synergistic action of graphite and a filler

In this section, we focus on the synergistic effects between graphite and single fillers. For instance, Majid Niaz Akhtar and colleagues studied the interaction between natural graphite and multi-walled carbon nanotubes (MWCNTs) in composite materials used for bipolar plates. [96] This synergy is attributed to the electron transfer phenomenon between natural graphite and MWCNT channels, ultimately enhancing the material's conductivity. Additionally, the addition of carbon nanotubes can also improve the bending performance of bipolar plates. This enhancement stems from the excellent mechanical strength and high aspect ratio of MWCNTs, while the interface adhesion between the matrix and MWCNTs also plays a crucial role in improving the bending performance. Furthermore, higher Young's modulus and hardness values enhance the mechanical strength of graphite and carbon nanotube composite materials. Additionally, carbon nanotubes can act as spacers or bridges between graphite particles, reducing contact resistance and increasing the interface area for charge transfer.

Aakash Masand [97] simultaneously used natural graphite (NG) and expanded graphite (EG) as conductivity fillers, and the results showed that at a constant 60% filler content, the composite material composed of 30% NG and 30% EG achieved the optimal conductivity of 416.66 S/cm, surpassing single conductivity filler materials.

Mathur R. B [98] and others studied the effect of carbon black as a filler in a three-component system. In the S-3 series of experiments, with the resin and natural graphite quantities held constant, carbon black replaced synthetic graphite. Figures 11(a-c) show the variations in density, resistivity, and Shore hardness of the plates with increasing carbon black content. As shown in Figure 11a, with the increase in carbon black content, the bulk density of the plate gradually decreases, evidently due to the lower bulk density of carbon black compared to synthetic graphite (<2.0 g/cm³). However, the reduction is minimal, amounting to only 2%.

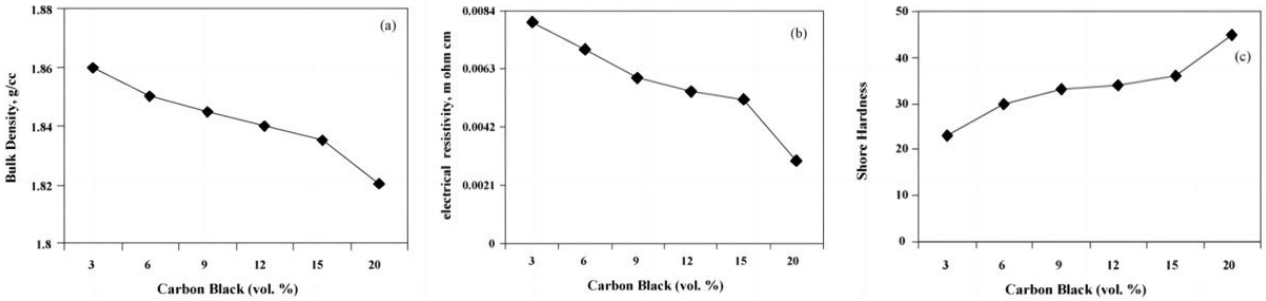

Figure 11. Variation in (a) bulk density, (b) electrical resistivity, and (c) shore hardness of composites bipolar plate with change in carbon black volume fraction.[98]

The advantages of adding carbon black are prominently manifested in the improvement of conductivity or reduction in the resistivity of the plate. As seen in Figure 11b, the resistivity of the plate decreases from 8 (3% CB) to 2.5 mcm (20% CB). This indicates that the small-sized carbon black particles (<1 μm) fill the gaps between natural graphite particles. By adding 20% CB to the mixture, the resistivity decreases by an order of magnitude. Figure 11c illustrates the variation in hardness of the composite plates with increasing carbon black content. The hardness value increases from 20 (5% CB) to 45 (20% CB). Since in this series of compositions, CB replaced an equivalent amount of SG, it can be concluded that the hardness values of SG and CB are the same. Experimental results show that adding carbon black can significantly enhance the conductivity of composite materials without compromising the density and hardness of the composite.

Anett Király and Ferenc Ronkay [99] utilized a melt blending method to prepare graphite-carbon black hybrid filler materials. Optical microscopy studies demonstrate that carbon black not only directly impacts the conductivity and mechanical properties of the composite material but also indirectly influences them due to its significant effect on the distribution of graphite. In enhancing conductivity, the two fillers exhibit a synergistic effect: the higher the graphite content, the more significant the improvement in conductivity with the addition of carbon black. In improving mechanical properties: when the graphite content is low, the hybrid composite material becomes more flexible due to the reduced size of graphite particles and the decreased average distance between particles, leading to better dispersion of graphite. However, when the carbon black content is high, graphite particles re-agglomerate, resulting in a significant increase in material stiffness and a decrease in impact strength.

3.2. Synergistic action of graphite and various fillers

For CBP, such as NG and EG, are the primary conductive fillers. The results also indicate that using multiple conductive fillers can further enhance the conductivity of CBP. The simultaneous use of multiple fillers is a trend in the development of composite materials. This approach provides significant performance enhancements compared to composite materials (CMs) containing only one type of filler. These improvements encompass various aspects such as conductivity, thermal conductivity, elastic properties (including strength, Young's modulus, and glass transition temperature), and mechanical losses. Additionally, a trade-off between electrical and mechanical performance should be considered. When the conductivity reaches a threshold, an excess of filler cannot further enhance the conductivity of CBP and may instead lead to a decrease in the material's mechanical properties. This section discusses the synergistic effects between graphite and multiple fillers, emphasizing their significance in enhancing material performance and expanding applications. A comprehensive investigation and understanding of these synergistic effects will yield valuable results.

For example, R.K. Gautam [100] et al. studied the synergistic effects of carbon fillers (including exfoliated graphite, carbon black, and graphite powder) in phenolic resin-based composite bipolar plates for PEM fuel cells. Their research indicates that the optimized filler composition of 35/5/3

(exfoliated graphite/carbon black/graphite powder) outperforms plates filled solely with exfoliated graphite in terms of battery performance.

Peretsetal [101] et al. examined the synergistic effects of graphite with two fillers, multi-walled carbon nanotubes (MWCNTs), and graphene nanoplatelets (GNPs) in the production of bipolar plates. The focus of the study was on enhancing the conductivity of the composite material. In low-viscosity polymers, the combination of the two conductive fillers exhibited a synergistic effect above the percolation threshold, resulting in conductivity improvements of up to 20 times. The reduction in electrical contact resistance in hybrid composite materials can be attributed to the thinning of the polymer layer between the increased number of filler particles participating in the conductive circuit.

The addition of carbon black not only improves the mechanical properties of the material but also enhances its conductivity and hardness. The lightweight and high-strength composite plates allow the use of thin plates with good strength, ensuring a lightweight fuel cell stack. R.B. Mathur [102] et al. used natural graphite, synthetic graphite, carbon fibers, and carbon black as reinforcement components, with phenolic resin as the binder matrix, fabricated using compression molding techniques. Through proper combinations and ratios, composite plates with a volume density of 1.8-1.90 g/cm³, resistivity of 0.002-0.007 cm, Shore hardness of ~65, bending strength of ~45 MPa, and bending modulus of ~12 GPa were produced. The performance of the developed composite plates was compared with commercially available bipolar plates. When used as bipolar plates for unit fuel cells at a current density of 1400 mA/cm², the power density reached ~500 mW/cm².

Shen Lie et al. incorporated two different-sized conductive fillers, carbon black (CB) and carbon fiber (CF), into a high-density polyethylene (HDPE) and polypropylene (PP) dual-component polymer system, producing a four-component conductive composite material. CB was selectively distributed within the HDPE matrix, forming a dual-continuous phase distribution with PP, establishing a dual-permeable conductive network structure. CF, with a high aspect ratio, was uniformly dispersed in the polymer phase and bridged multiple phase regions, aiding in the formation of the conductive network. Electrical characterization indicated a significant decrease in volume resistivity by 1 to 5 orders of magnitude in the four-component system compared to the three-component composite system. The presence of a dual-permeable conductive network effectively suppressed the negative temperature coefficient (NTC) effect and enhanced cycling stability.

Sirawit Witpathomwong [103] et al. developed composite materials for proton exchange membrane fuel cell (PEMFC) bipolar plates by filling graphite, graphene, and carbon nanotubes. In the composite material filled with 2% carbon nanotubes, the through-plane thermal conductivity reached as high as 21.3 W/mK, 44 times higher than the thermal conductivity of other polymer composite bipolar plate systems. Furthermore, the study found that the maximum graphite content in polyphenylene benzobisoxazole-based composite materials filled with graphite was 84 wt% (74 vol%), exceeding this content led to a slight decrease in the actual density of the composite material compared to the theoretical density. By filling 7.5 wt% graphene and varying the carbon nanotube content, it was observed that the actual density of the composite material decreased at 2 wt% carbon nanotube content, possibly due to decreased wettability of the matrix leading to the formation of some voids. The fracture surface of the graphite/graphene/carbon nanotube/polyphenylene benzobisoxazole-based composite material exhibited excellent dispersion of various fillers in the polymer matrix, attributed to the low melting viscosity of the benzobisoxazole resin and its good wetting properties. The thermal conductivity, electrical conductivity, flexural strength, and other relevant properties of the composite material filled with carbon nanotubes met the requirements of the U.S. Department of Energy, making this composite material an ideal choice for PEMFC bipolar plates.

Yao [104] et al. selected three types of graphite, SG, NG, and EG, as conductive fillers and used PF as the binder to prepare composite bipolar plates. The study revealed that the PF/EG composite bipolar plates prepared with EG as the conductive filler exhibited the best performance, with an electrical conductivity of 182 S/cm and a high bending strength of up to 109 MPa at an EG content of 80%.

Due to the weak van der Waals forces in the graphite interlayer, it can reduce the in-plane conductivity. Therefore, many studies use nano-sized particles to form a conductive path by filling the void spaces within the graphite interlayer, which can enhance the through-plane conductivity of composite BP. Du et al. prepared carbon-filled epoxy composites using EG and CB to achieve a synergistic combination of in-plane and through-plane conductivity. Embedding carbon black powder in the EG layer of the epoxy resin reduced the conductivity of the composite BP. They reported an in-plane conductivity of the composite BP exceeding 1000 S/cm, attributed to the synergistic effects of EG and CB, as shown in Figure 12. Adloo et al. investigated the impact of conductive carbon fillers (such as graphite, graphene, and CB) on the manufacture of polypropylene BP. The results demonstrated a significant influence of adding carbon particles on the conductivity, reaching a conductivity of 104.63 S/cm. [105-107]

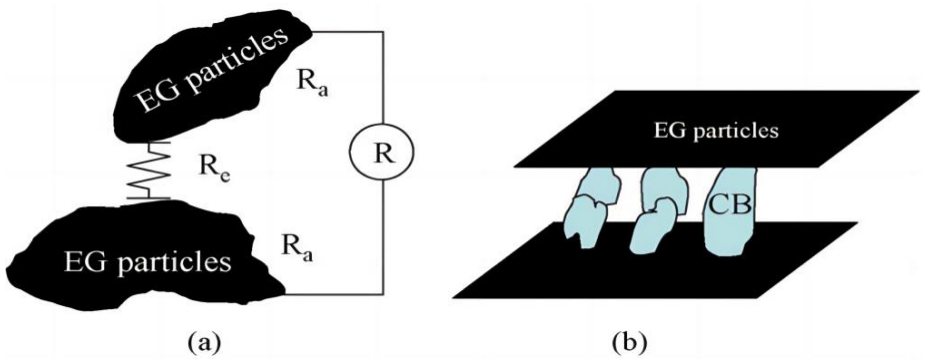

Figure 12. Schematic of (a) how various resistances act in series and define overall resistance R and (b) CB particles placed between two graphite platelets to help reduce value of [106]

4. Dual Percolation Effects and Their Applications

The application of the dual percolation effect in resins typically involves the preparation of conductive composite materials, especially critical components of electrochemical devices like bipolar plates. This effect is achieved by introducing two incompatible polymers into the matrix. Since the matrix is insulating, the composite material exhibits high resistivity when the filler concentration is below the percolation threshold. As the filler concentration increases, conductive pathways begin to form, leading to a decrease in resistivity. The region where resistivity decreases rapidly is known as the percolation region (as shown in Figure 13). Subsequently, as the conductive network is well established or the filler concentration in the system is close to saturation, further increasing the filler content results in only a minimal decrease in resistivity.

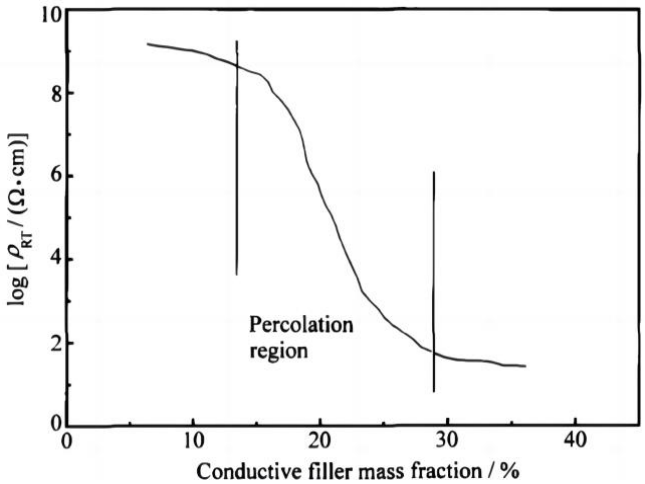

Figure 13. Schematic drawing of room-temperature resistivity vs filler mass fraction of a conductive polymer composite

The preparation of conductive composite materials with a dual percolation conductive network requires significantly high concentrations of particle loading. Yu et al. reported the percolation threshold of conductivity of MWCNTs in polyphenylene sulfide (PPS) [108]. They reported that the viscosity sharply increased above 2wt% MWCNTs, while the electrical conductivity of the composite bipolar plate containing 2wt% MWCNTs improved at 2wt% MWCNTs (1.12×10^5 Ω·cm). The results indicate that high particle concentrations increase melt viscosity and reduce the mechanical properties of the composite material.

Recently, it has been reported that multiphase polymer blends can reduce particle loading, as the conductive filler can selectively reside between two phases or within one of the polymer phases (see Figure 14) [109], [110], [113]. When the dispersed conductive filler reaches the percolation threshold, a continuous structure of the conductive filler forms in the polymer, known as the dual percolation threshold. The dual percolation method allows for the formation of conductive pathways in composite materials with a small amount of conductive filler [114], [115].

Alo et al. [116] studied the synergistic effect of PP/epoxy resin/GR composite materials in a co-continuous morphology. They introduced CB as an additional minor filler.

Figure 15 compares the electrical conductivity of two composite materials: PP/epoxy resin/73 wt.% GR/7 wt.% CB (total filler 80 wt%) and PP/epoxy resin/80GR (80 wt% GR without CB). The results indicate an increase of 30% in in-plane electrical conductivity for the former and a 165% increase for the latter. This enhancement in conductivity is attributed to the CB particles filling the spaces between the larger GR particles, creating additional electrical pathways in the composite material.

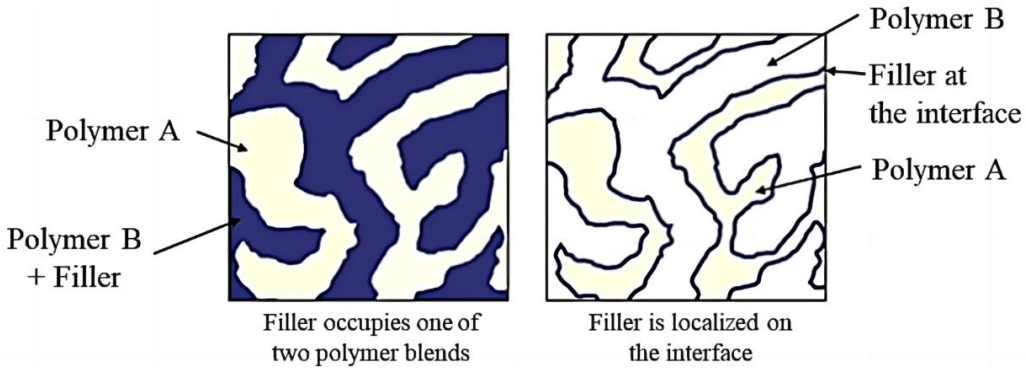

Figure 14. Schematic of filler distribution in double percolation theory [114]

For multiphase blend systems, the electrical performance of the system is ultimately controlled by the formed conductive network, closely related to the phase morphology and filler dispersion. Yui et al. [117] observed the behavior of CB in immiscible polyethylene HDPE/polypropylene (PP) blends. They mixed HDPE/PP using a twin-screw extruder at 240°C/200rpm. The blended polymers were pelletized. Subsequently, thick composite materials were prepared through hot pressing and injection ion molding. In the CB-reinforced HDPE/PP blend, the composite material consisted of three phases: CB-HDPE composite, HDPE, and PP phases, as the CB particles selectively resided in the HDPE phase. Nguyen et al. [118] observed a similar trend for PET/CB, PVDF/CB, and co-continuous (PET/PVDF)/CB composite materials. The co-continuous PET/PVDF morphology exhibited higher conductivity as CB primarily positioned within the PET phase. Additionally, reducing the PET ratio from 60 wt% to 50 wt% in the PVDF/PET blend enhanced the conductivity network through a decrease in surface resistivity. However, decreasing the PET concentration to 30 wt% resulted in an increase in surface resistivity, indicating the disruption of the PVDF/PET morphology and the dispersion of PET within the PVDF matrix.

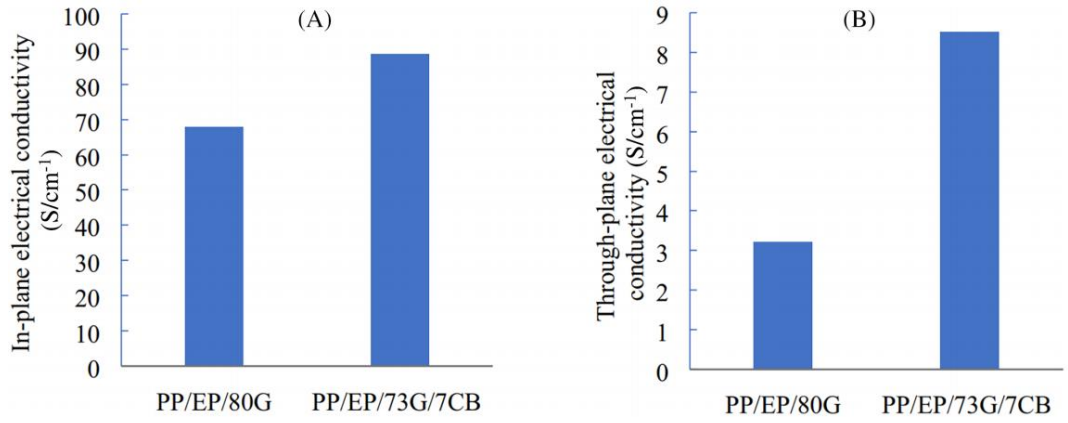

Figure 15. Electrical conductivity of PP/EP/80 wt.%GR and PP/EP/73 wt.%GR/7 wt.%CB composites in (A)in-plane direction;(B)through-plane direction (CB,carbon black; EP, epoxy; G,graphite; PP, polypropylene). [116]

Negative temperature coefficient (NTC) is a phenomenon where the resistance decreases with increasing temperature in certain materials. Through the dual percolation effect, it is possible to reduce the percolation threshold and room temperature resistivity of materials, suppressing the NTC phenomenon and ensuring greater stability in the electrical properties within a specific temperature range. Simultaneously, by incorporating two different-sized conductive fillers in the polymer matrix, the room temperature resistance can be reduced, enhancing the Positive Temperature Coefficient (PTC) strength.

Overall, dual infiltration behavior provides an effective approach to enhancing the electrical performance stability of composite materials by optimizing the conductivity network structure, reducing resistance, and suppressing negative temperature coefficient phenomena. This research direction holds significant potential importance for material design and applications in areas such as electronics, batteries, and electric vehicles.

5. Conclusion

Proton Exchange Membrane Fuel Cells (PEMFCs) are promising power generation devices known for their high energy conversion efficiency, quick cold start at low temperatures, and lack of pollution. Among the core components of PEMFCs, the bipolar plates play a significant role in influencing the overall performance of PEMFCs. This article provides a brief overview of the research progress on polymer/conductive filler composite bipolar plates for PEMFCs, summarizing the current research directions and relevant properties of these materials.

Currently, thermosetting resins such as phenolic resins and epoxy resins are the primary polymers used in the research of polymer/conductive filler composite bipolar plates for PEMFCs, with fewer studies focusing on thermoplastic resins. Graphite is the main conductive filler used, but the conductivity of composite bipolar plates made solely with graphite as the conductive filler is limited. Incorporating appropriate amounts of carbon black, carbon fibers, carbon nanotubes, and graphene as conductive additives can significantly enhance the comprehensive performance of composite bipolar plates. Therefore, the future trend in research for polymer composite bipolar plates involves synergistic enhancement through multiple conductive fillers, emphasizing the importance of understanding the behavior of different carbon-based fillers in specific organic matrices due to infiltration phenomena. Below are recommendations regarding material selection and manufacturing methods:

1) To reduce the infiltration threshold and improve the conductivity of composite materials, maximize the aspect ratio of the conductive particles.

2) Elongated particles often exhibit higher conductivity compared to spherical fillers. In this context, graphite nanoplates and carbon nanotubes play a dominant role in enhancing the electrical

performance of composite bipolar plates. Similarly, flake graphite particles are more preferred over spherical graphite particles.

3) The particle size should be maximized for a given content of conductive filler.

4) The influence of carbon fibers on the electrical conductivity of composite bipolar plates is closely related to their orientation, hence closely tied to the manufacturing method.

5) A small amount of carbon black can enhance the electrical performance of composite bipolar plates by forming additional conductive networks between graphite particles.

6) Carbon nanotubes exhibit stronger conductivity than carbon black, and their effectiveness when incorporated into a polymer matrix depends on their thorough dispersion. Functionalization can be used to reduce the tendency of nanotubes to cluster, hindering their uniform distribution and lowering conductivity.

7) Poor dispersibility favors the aggregation of conductive particles, thereby reducing the formation of infiltration networks and the overall conductivity of composite materials.

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