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REVIEW

Review of Conductive Polymer Coatings for Metallic Bipolar Plates in Proton Exchange Membrane Fuel Cells

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ABSTRACT

Proton exchange membrane fuel cells (PEMFCs) have gained increasing interests as promising power sources due to their ability to convert hydrogen and oxygen directly into electricity with high efficiency and zero greenhouse gas emissions. Bipolar plates (BPs) are considered as a critical component of PEMFCs, serving to collect current, separate gases, distribute the flow field, and conduct heat. This paper reviews the technical status and advancements in BP materials, with special focus on strategies for enhancing interfacial contact resistance (ICR) and corrosion resistance through conductive polymer (CP) coatings. First, commonly used BP materials in PEMFCs are summarized. Then, the advantages and limitations of various coatings for metallic BPs are discussed. Finally, recent progress in CP coatings for metallic BPs, aimed at achieving high corrosion resistance and low ICR, is comprehensively reviewed.

KEYWORDS

PEMFC; bipolar plate; conductive polymer coating; polypyrrole; polyaniline

1 Introduction

In recent years, the push to develop clean energy solutions has been driven by the urgent need to mitigate environmental pollution and address the depletion of traditional energy sources, challenges intensified by population growth and societal development [1,2]. Renewable and clean energy sources, including hydrogen, solar, wind, geothermal and biomass, have garnered considerable attention for their potential as sustainable alternatives to non-renewable fossil fuels [3–5]. However, most renewable sources, aside from hydrogen, exhibit significant volatility and unpredictability, which constrain their feasibility for consistent daily use [6]. This limitation can be mitigated by converting these renewable resources into hydrogen, which can be stored and used on demand [7]. Thus, research into efficient hydrogen production, storage, transportation and utilization methods is both timely and essential [8].

Proton exchange membrane fuel cells (PEMFCs) stand out as one of the most promising technologies for hydrogen utilization, with the potential to serve as viable alternative to internal combustion engines in future vehicles due to their rapid start-up, near-zero carbon emissions, long service life, and high-power density [9–11]. PEMFCs, known for their high efficiency and modular design, have seen expanding applications in various modes of transportation, including cars, buses, motorcycles, ships, submarines and aircraft [12]. This versatility underscores their strong potential as a clean and reliable power source for a wide range of high-energy-demand applications across multiple market sectors [13]. As illustrated in Fig. 1, key components of PEMFCs include bipolar plates (BPs), gas diffusion layers (GDLs), catalyst layers (CLs) and proton exchange membranes (PEMs). Notably, BPs alone account for approximately 80% of the total weight, 50%–65% of the total volume and 30%–40% of the total cost of a PEMFC system [14]. Consequently, the large-scale application of PEMFCs requires hinges on advancing the development of BPs that are both cost-effective and high-performing [15].

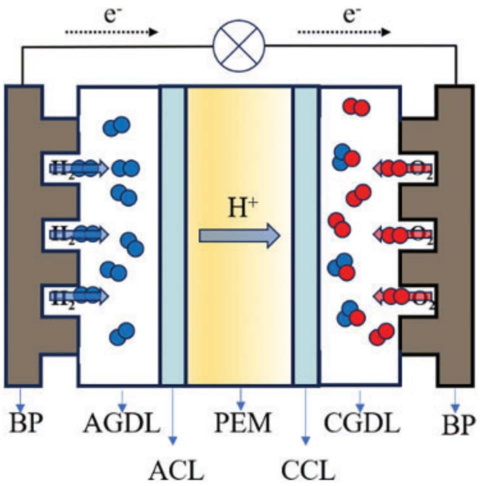

Figure 1: Schematic diagram of PEMFC (AGDL: Anode gas diffusion layer; CGDL: Cathode gas diffusion layer; ACL: Anode catalyst layer; CCL: Cathode catalyst layer; PEM: Polymer electrolyte membrane; BP: Bipolar plate)

The objective of this work is to provide a comprehensive overview of the development of BPs in PEMFCs, with a particular focus on the exploration of protective coatings for conductive polymers (CPs) designed to enhancing electrical conductivity and corrosion resistance. This review is structured as follows: (a) Section 2 outlines the technical requirements of BPs and examines the material characteristics of various BP types. (b) Section 3 summarizes typical coatings applied to modify metallic BPs, highlighting their merits and limitations in practical applications. (c) Section 4 delves into the CPs, covering synthesis methods, corrosion mechanisms and recent advancements in both intrinsic conducting polymers (ICPs) and non-intrinsic conducting polymers (NICPs). (d) Section 5 introduces the life cycle assessment and environmental impact of CP coatings for fuel cell applications. Finally, conclusions and future perspectives are presented in Section 6.

2 The Overview of Materials for BPs

The essential roles of BPs in PEMFCs are as follows: (1) providing conductive pathways for collecting and conducting the output current; (2) isolating the reactants at the cathode and anode; (3) separating gases and facilitating water discharge by managing gas and liquid flows; (4) connecting individual cells and supporting the stack structure to reinforce the membrane; (5) dissipating heat [16,17]. To effectively fulfill these functions, ideal BPs must possess excellent mechanical strength, machinability, corrosion resistance, chemical stability, electrical conductivity, thermal conductivity and gas impermeability [18,19]. Table 1 presents the specific technical parameters targeted for bipolar plates, as outlined by the Department of Energy (DOE), with milestone goals set for 2020 and 2025, respectively [19–21].

Table 1: DOE targets for BP performance [20,21]

Characteristics	Units	2025 DOE targets	2020 DOE targets
Electrical	S cm^{-1}	>100	100
Conductivity areal specific resistance	Ω cm^2	<0.01	0.01
H_2 permeability	cm^3 s^{-1} cm^{-2}	2×10^{-6}	1.3×10^{-14}
Thermal conductivity	W m^{-1} K^{-1}	/	10
Corrosion, anode corrosion, cathode	μA cm^{-2}	<1 and no active peak	<1 and no active peak
Lifespan	hours	8000	5000
Plate weight	kg kW^{-1}	0.18	0.4
Cost	\$ kW^{-1}	2	3
Flexural strength	MPa	>40	25

In the development history of PEMFC BP materials, graphite, metallic and composite BPs have emerged as primary candidates due to their distinctive properties [22]. Table 2 provides an overview of the main advantages and disadvantages of these three types of materials [17,20,23–26]. Traditionally, graphite has been favored for BPs due to its high electrical conductivity and excellent chemical stability. It offers low bulk resistivity and robust corrosion resistance, attributes desirable for long-term PEMFC applications. However, graphite's inherent brittleness, high cost and challenging machinability necessitate greater thickness for structural integrity, leading to increased stack weight and volume, which limits its commercial viability, particularly for transportation applications [27,28]. Composite BPs present improvements over graphite in terms of corrosion resistance and mechanical strength. However, composite BPs typically have lower electrical conductivity than metallic BPs, and their complex manufacturing processes hinder largescale commercial application [29]. In contrast, metallic BPs, with their favorable mechanical properties and cost-effectiveness, are better suited for largescale production and supporting the membrane-electrode assembly [30,31]. Metallic BPs also offer key advantages, such as low gas permeability and excellent electrical and thermal conductivity, positioning them as promising materials for PEMFC BPs [32,33].

Table 2: Advantages and disadvantages of different BP materials [17,20,23–26]

Materials	Advantages	Disadvantages
Graphite	Excellent corrosion resistance and conductivity	Low mechanical strength, poor machinability
Composite material	Good corrosion resistance, high strength, compact size, lightweight	Poor conductivity, high cost, not suitable for large-scale manufacturing
Metal	Excellent thermal and electrical conductivity, high strength, low permeability, low cost	Prone to corrosion (which degrades conductivity and can lead to metal ion contamination of catalysts and membrane electrodes)

Various commercial metals, including stainless steels, titanium, nickel, copper and aluminum alloys, are currently used as metallic BPs [15,34]. Stainless steel is frequently chosen for its ease of processing, robust mechanical properties and moderate chemical stability [35]. Nonetheless, its high density poses a drawback for weight-sensitive transportation applications, and it typically requires protective coatings to prevent corrosion in the acidic environment of PEMFCs. Titanium, a lighter and more corrosion-resistant option, enhances durability in high-temperature operating conditions and reduces stack weight, but its high cost and relatively low conductivity limit its largescale applications [36,37]. Aluminium, while lightweight and cost-effective, suffers from low corrosion resistance and also requires surface coatings to prevent degradation in acidic conditions [38]. Nickel and copper provide high thermal and electrical conductivity as well as solid mechanical strength, but their higher densities and susceptibility to corrosion in PEMFC conditions pose additional challenges [39,40]. To provide a clear comparison of the performance of different materials as PEMFC BPs, Fig. 2 presents the corrosion rates, ICR values, costs and mechanical strength of graphite, composite materials and various metallic materials. As shown in Fig. 2, while metallic BPs generally demonstrate reliable corrosion resistance due to passivation properties, they still face significant challenges in PEMFC environments, where passive films on their surfaces are vulnerable to break down [17,41]. Corrosion of BPs can degrade PEMFC performance by contaminating the membrane and catalyst with dissolved ions, thus comprising structural stability [16,42,43]. Additionally, the passive films on the BP surfaces contribute to the increase of interfacial contact resistance (ICR), which reduces the efficiency and durability of the PEMFC stack [44]. Considering that both corrosion and ICR are directly related to the surface characteristics of BPs, surface modification technologies have been naturally adopted to address these issues in metallic BPs [45,46].

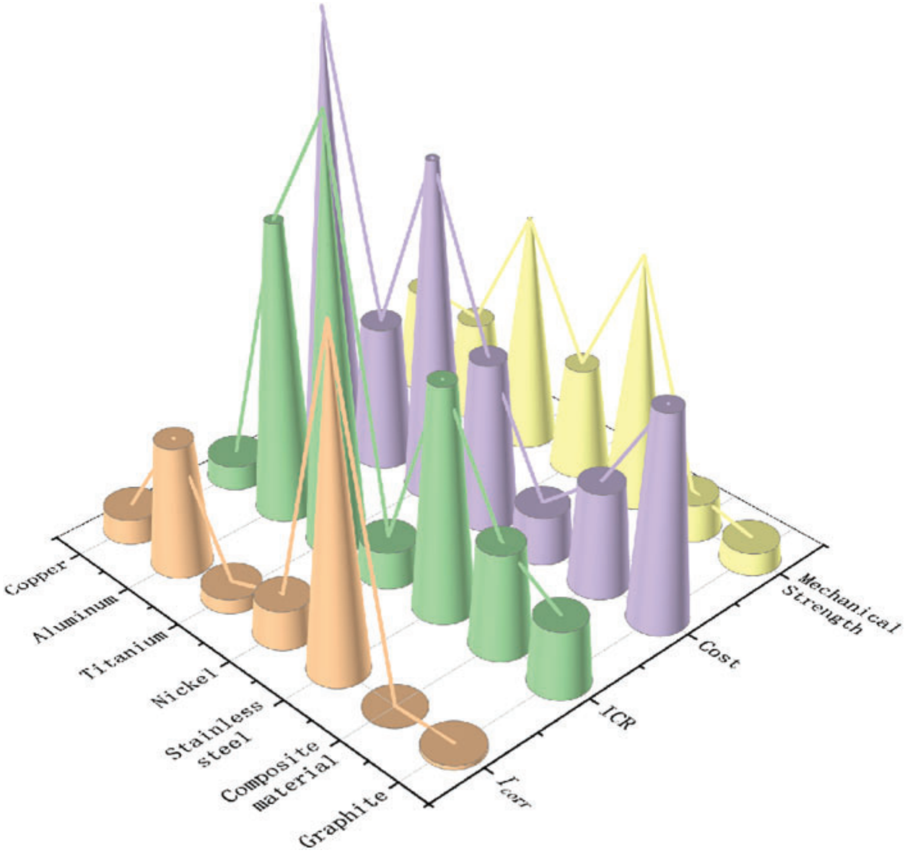

Figure 2: Comparison of performance of different BP materials for PEMFCs [31,47–50]

3 Coatings for Metallic BPs

A variety of coatings have been proposed to enhance the performance of metallic BPs in PEMFCs by improving corrosion resistance and reducing ICR. The main materials considered suitable for protective coatings on metallic BPs include: (1) noble metals, (2) metal nitrides, (3) metal oxides, (4) metal carbides, (5) carbon-based materials and (6) conductive polymers (CPs) [17,46].

Table 3 summarizes typical investigations on protective coatings for metallic BPs, highlighting their processing methods, corrosion resistance, ICR, as well as their merits and limitations in practical applications [46,49,51–63]. While each coating type offers distinct benefits, they also present specific challenges. Among these, CPs stand out as particularly promising protective barriers for BPs owing to their excellent corrosion resistance. However, most CPs exhibit relatively high ICR when interfacing with the gas diffusion layer, which can adversely affect overall performance [49,64–66]. To address this limitation, innovative strategies aiming at improving the electrical conductivity and durability of CPs are essential for enabling their large-scale commercial application. The following section provides a comprehensive overview of CPs focusing on critical technologies, underlying mechanisms and key research advancements, as well as future potential for use in PEMFC BPs.

Table 3: Some typical studies on coatings for metallic BPs and their characteristics [46,49,51–63]

Coatings	BPs	Processing methods	Electrolytes	i (μA cm^{-2})	ICR (mΩ cm^2)	Coating types	Characteristics	Ref.
Ag	316L SS	Ion implantation	0.5 M H_2SO_4 + 2 ppm HF, 80°C	0.7	78.8	Inert metals	Excellent property, but expensive for large-scale applications	[46]
Au	316L SS	Ion implantation	0.5 M H_2SO_4, 80°C	0.2	0.9			[51]
CrN	316L SS	Arc ion plating	0.5 M H_2SO_4 + 2 ppm HF, 80°C	0.03	23	Metal nitrides	Excellent property, but expensive for large-scale applications Low cost, good conductivity, but difficult to prevent the invasion of corrosion fluid	[52]
TiN	316L SS	Cathode arc ion plating	0.5 M H_2SO_4 + 2 ppm HF, 80°C	40	10			[52]
1.86% Al-CrN	316L SS	Magnetron sputter ion plating	0.5 M H_2SO_4 + 5 ppm HF, 70°C	0.021	5.1			[53]
$Nb_{0.04}$-TiO_2	316L SS	Sol-gel method	0.1 M H_2SO_4, 80°C	0.13	38	Metal oxides	Low cost, good conductivity, but difficult to prevent the invasion of corrosion fluid Good corrosion resistance, but poor conductivity	[54]
1.3% F^-SnO_x	316 SS	Chemical vapor deposition	1 M H_2SO_4 + 2 ppm HF, 70°C	–	74			[55]
Ti_4O_7	Ti	High-temperature roasting	0.1 M H_2SO_4 + 2 ppm HF, 70°C	–	7.61			[56]

(Continued)

Table 3 (continued)

Coatings	BPs	Processing methods	Electrolytes	i (μA cm^{-2})	ICR (mΩ cm^2)	Coating types	Characteristics	Ref.
$Cr_{0.75}C_5$	316L SS	Magnetron sputter ion plating	0.5 M H_2SO_4 5 ppm HF, 70°C	1.046	1.4	Metal carbides	Good corrosion resistance, but poor conductivity Excellent property, but complex process and high equipment requirements	[57]
Nb-C	304 SS	Double glow plasma alloying	0.5 M H_2SO_4 + 2 ppm HF, 80°C	0.02	8.47			[58]
TiC	304 SS	Micro-arc alloying	1 M H_2SO_4, room temperature	3.7	–			[59]
a-C	316L SS	Magnetron sputter ion plating	0.5 M H_2SO_4 + 0.1 ppm HF, 80°C	<0.05	2.44	Carbon based material	Excellent property, but complex process and high equipment requirements	[60]
a-C	316L SS	Magnetron sputter	0.5 M H_2SO_4 + 5 ppm HF, 80°C	0.0075	2.91			[61]
Graphene	304 SS	Chemical vapor deposition	3.5 wt% NaCl, room temperature	–	36			[62]
CSA-PPy	304 SS	Electrochemical deposition	0.1 M H_2SO_4, room temperature	<0.1	5.5	CPs	Poor adhesion	[63]
PANI	304 SS	Electrochemical deposition	0.5 M H_2SO_4 + 2 ppm HF, 80°C	0.23	–			[49]

4 CP Coatings

The use of CPs as corrosion barriers began with DeBerry's pioneering work in 1985, when polyaniline (PANI) films were applied on stainless steel to prevent corrosion [67]. Since then, CPs have shown immense potential for corrosion protection, thanks to their effective barrier properties, passivation and corrosion inhibition effects [66,68–70]. In addition, their intrinsic conductivity, in conjunction with excellent chemical inertness, makes CPs promising candidates for surface coatings on metallic BPs [66,71]. These CPs, commonly referred to as intrinsically conductive polymers (ICPs), derive their conductivity from their inherent conductivity.

Initially, research on CPs, particularly ICPs such as PANI and polypyrrole (PPY), focused on optimizing basic barrier properties and corrosion protection by adjusting electrochemical deposition parameters and conducting tests in acidic environments. However, as the field evolved, attention shifted toward improving the long-term stability of these coatings. Advances such as doping and the simulation of PEMFC operating conditions have significantly enhanced their performance. Despite these improvements, ICPs often suffer from poor adhesion to metallic BPs due to their limited polar functional groups, which remains a significant barrier to practical application [66,72].

To address these challenges, researchers have developed CP composites that combine high conductivity with improved corrosion resistance. Strategies include: (1) Double-layer composite coatings: these feature an outer layer for corrosion protection and an inner layer to enhance conductivity. (2) Incorporation of conductive nanofillers: materials like carbon nanotubes (CNTs) and graphene improve both the performance and durability of CP coatings.

In contrast to ICPs, non-intrinsically conductive polymers (NICPs) offer an alternative solution. by embedding conducting fillers into non-conductive polymer matrices, NICPs enhance adhesion to metallic substrates while maintaining satisfactory conductivity. The conducting fillers facilitate efficient electron transport, improving electrical performance [66,73]. Besides, double-layer or multi-layer coatings have been developed to further augment the corrosion resistance and optimize overall functionality of CP coatings [74].

Current research is dedicated to advancing preparation methods, elucidating corrosion mechanisms, and enhancing the electrical conductivity of CP coatings for metallic BPs. However, several challenges persist: (1) Corrosion resistance: CPs must endure the highly acidic and humid conditions typical of PEMFC environments without degradation. (2) Conductivity: high ICR can impede electron transfer efficiency, reducing overall fuel cell performance. (3) Long-term stability: continuous exposure to fluctuating temperatures and electrochemical cycling can cause delamination or breakdown of the polymer structure.

The following section delves into the research progress of CP coatings on metallic BPs, focusing on ICPs, NICPs, and multi-layer CP coatings. It discusses fabrication methods, recent advancements, and the limitations associated with each type, providing a comprehensive overview of their development and application.

4.1 ICP Coatings

ICP coatings are generally synthesized using either chemical or electrochemical methods [75]. Chemical synthesis, mainly involving oxidative polymerization, uses an oxidizing agent to initiate polymerization by oxidizing the monomer and forming a polymer coating [76]. However, this method often faces challenges such as complex synthesis process, difficult reaction control and potential environmental pollution from by-products, restricting its practical application. In contrast, electrochemical methods, such as the constant current method and cyclic voltammetry (CV), achieve polymerization through electron charge reactions [77]. Comparatively, the electrochemical methods offer simpler synthesis process, precise reaction control and greater suitability for practical applications, making them the focus of ICP research [67,75–78]. However, issues such as heterogeneous nucleation during the electrodeposition process and poor adhesion can reduce the corrosion resistance and conductivity of these coatings [66,79]. Therefore, further advancements are essential to enhance the quality and performance of electrodeposited ICP coatings.

The protective mechanism of CPs primarily lies in their function as a physical barrier that blocks the penetration of corrosive ions, along with anodic protection achieved by passivating the substrates. The corrosion behavior of ICP coatings is strongly associated with their specific types [66,69]. Among the various ICPs, PANI and PPy have been the most extensively studied for corrosion protection [66]. The following sections will discuss the advancements in these ICP coatings, focusing on their corrosion protection mechanisms and strategies for performance enhancement.

4.1.1 PANI Based ICPs Coatings

PANI is a widely recognized conductive polymer known for its impressive thermal stability, environmental durability, corrosion resistance, and notable optical and electrical properties [68]. As shown in Fig. 3, PANI exists in three distinct oxidation states depending on the value of x in its basic structure: (1) leucoemeraldine (LE) in the fully reduced state ($x = 0$); (2) emeraldine (EB) in the half-oxidized state ($x = 0.5$); (3) pernigraniline (PE) in the fully oxidized state ($x = 1$) [68,80]. While undoped PANI is nearly insulating, the EB form achieves conductivity by various doping strategies, including charge transfer group doping, proton acid doping and charge injection at metallic/semiconductor interfaces [81,82]. The conductivity makes PANI an attractive option for CP coatings on BPs. Nevertheless, ensuring adequate corrosion resistance of PANI remains a critical consideration for its effective application [83].

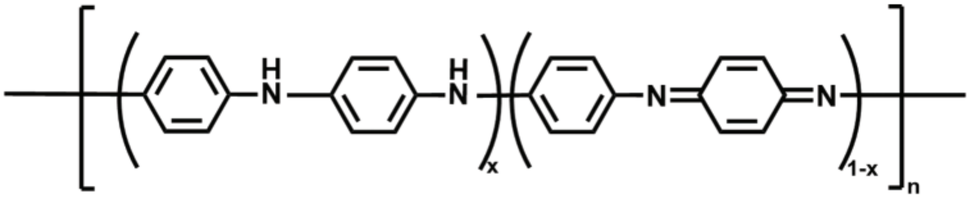

Figure 3: Molecular structure of PANI [66,78]

PANI serves as an effective anti-corrosion barrier by preventing the penetration of water, oxygen, and corrosive ions (e.g., Cl^-), thereby protecting the BP surfaces from direct exposure to these corrosive agents [84]. Additionally, PANI provides sustained anodic protection due to its unique redox properties facilitated by doping [85]. When PANI is in contact with a passivating metal substrate, its higher potential compared to the metal substrate initiates anodic polarization in the presence of water and oxygen [66,85]. This reaction encourages the formation of a passivating oxide layer on the metal surface, significantly reducing the corrosion rate. Simultaneously, PANI undergoes a reversible reduction to its LE form. In an oxidative electrolyte solution, the reduced PANI re-oxidizes back to its EB form, maintaining continuous anodic protection for the metal substrate [86]. The reversible redox cycle of PANI can be described by the following Eqs. (1) and (2):

Reduction reaction of intermediate PANI:

$$(1/n)\,\mathrm{M} + (1/m)\,\mathrm{ES}^{m+} + (y/n)\,\mathrm{H_2O} \rightarrow (1/n)\,\mathrm{M}\,(\mathrm{OH})_y^{(n-y)+} + (1/m)\,\mathrm{EB}^0 + (y/n)\mathrm{H}^+ \tag{1}$$

Reoxidation of reduced PANI by dissolved oxygen:

$$(m/4)\,\mathrm{O_2} + m\mathrm{H}^+ + \mathrm{EB}^0 \rightarrow \mathrm{ES}^{m+} + (2/m)\,\mathrm{H_2O} \tag{2}$$

Wessling et al. found that doping PANI enables the formation of an oxide layer up to 1 μm thick on metal surfaces, providing effective passivates. Remarkably, when the passivation layer is damaged, PANI exhibits a "self-healing" ability by repairing and maintaining the integrity of the protective layer over time. This self-healing property significantly enhances the long-term anti-corrosion performance of PANI-coated metal surfaces [87].

To satisfy the demanding requirements of BPs in harsh environments, further improvements in corrosion resistance and conductivity are necessary due to the intrinsic deficiencies of PANI. Doping modification has proven to be an effective strategy for achieving these improvements [88,89]. The reversible doping/de-doping properties of PANI allow for the use of a wide range of dopants, including inorganic acids, organic anions and inorganics, to optimize its performance [67,90].

Common inorganic acids used for doping PANI include sulfuric acid (H_2SO_4) and hydrochloric acid (HCl) [91–93]. For instance, Le et al. successfully deposited H_2SO_4-doped PANI on 316L stainless steel (SS) by CV in a solution with 0.1 M aniline and 0.1 M H_2SO_4. The resulting coating exhibits corrosion resistance and ICR values meeting the recommended thresholds for BPs in PEMFCs (current densities <16 μA cm^2 and ICR <10 mΩ cm^2) [91]. Gao et al. synthesized phosphomolybdic acid-doped PANI on 304 SS through electropolymerization, where the phosphomolybdic acid filled the pores in the PANI layer by forming oxides spontaneously, enhancing corrosion resistance [92]. Additionally, Cindrella et al. demonstrated that doping PANI with organic acids, such as camphor sulfonic acid (CSA) and p-toluene sulfonic acid (pTSA), outperformed HCl doping under low-humidity fuel cell conditions. This improvement was attributed to the larger organic anions ($p\text{-}TS^-$ and CS^-), which resisted de-doping and repelled corrosive anions, offering superior performance compared to smaller inorganic anions like Cl^- [69,93,94]. Similarly, Kamaraj et al. synthesized benzoate-doped PANI by dispersing and stirring it in 1 M NH_4OH, with the resulting iron-dopant salt promoting metal substrate passivation and improving corrosion resistance [95]. Ren et al. directly electrodeposited PANI doped with dodecylbenzesulfonate anions (DBS^-) on 304 SS in a solution containing 0.3 M aniline and 0.15 M sodium dodecylbenzesulfonate (SDBS). The DBS^- doped PANI coating effectively blocked corrosive species and acted as a catalytic oxidizer, thereby maintaining the passive state of 304 SS substrate in acidic solutions containing Cl^- or SO_4^{2-} ions, offering exceptional corrosion protection [96].

Reinforcing PANI coatings with secondary phases has also proven effective in enhancing their properties. This strategy integrates inorganic particles or carbon-based materials into the polymer matrix to improve conductivity and corrosion resistance [66,96–99]. For instance, Deyab et al. developed PANI/Zn-Pr composite coatings on 303 SS to increase the output density of PEMFC cells. They found that incorporating 1% Zn-Pr particles into the PANI matrix effectively filled the pores, providing excellent anti-corrosion performance in the fuel cell electrolyte environment [98]. Similarly, Sharma et al. created a PANI-TiN composite coating by doping PANI with TiN particles, achieving reductions in ICR value to 32 mΩ cm^2 and corrosion current density to approximately 0.5 μA cm^{-2} [99], demonstrating significant improvements in conductivity and corrosion resistance.

Conductive dopants not only reduce microspores and microcracks in polymer coatings but also provide electronic transmission channels, thereby improving the conductivity and corrosion resistance performance simultaneously [100]. Therefore, tremendous efforts have been dedicated to incorporating inorganic carbon materials as dopants to modify CPs, given the high specific surface area, electrical conductivity, and chemical stability of carbon materials, such as graphene (G), carbon nanotubes (CNTs) and carbon nanofibers [101–104]. For instance, Yu et al. reported a PANI/CNTs composite coating on 316 SS BPs for PEMFCs by pulse electrodeposition. The CNTs and pulse method improved the compactness, corrosion resistance and electrical conductivity of PANI coating, reducing its current density to 1.9 μA·cm^{-2} and lowering the ICR by 35% compared with pure PANI coating [102]. Similarly, Deyab et al. also discovered that the CNTs in the PANI matrix not only improve charge transfer by serving as conductive bridges between quinonoid rings, but also reduce the permeability of the PANI coating to oxygen and water, further boosting corrosion resistance [103]. Qi et al. developed waterborne PANI/G composite coatings, achieving an ICR of 10 $m\Omega \cdot cm^2$ and a corrosion current density (i_{corr}) of 0.88 $\mu A\ cm^{-2}$, meeting the requirements for metallic BPs in PEMFCs [104]. Qiu et al. investigated a PANI-GO (graphene oxide) composite coating using pulse current deposition technology. They claimed that the inclusion of GO and optimized deposition parameters contribute to a highly effective, low porosity PANI-GO corrosion-resistant coating [105].

4.1.2 PPy Based ICPs Coatings

PPy is another representative ICP known for its excellent conductivity, chemical stability, mechanical properties and non-toxicity [106]. PPy is typically synthesized by oxidative polymerization of monomer pyrrole, using methods such as chemical polymerization, electrochemical polymerization and chemical vapor deposition (CVD) [107]. PPy chains are generally formed through α-α bonding between pyrrole units, with chain growth terminated by end-blocking groups [107,108]. As shown in Fig. 4, PPy can be classified into two dissimilar types according to conductivity: (1) the non-conducting benzoid structure; (2) the conducting quinoid structure. In this form, the presence of conjugated double bonds enables electron flow, contributing to conductivity [72].

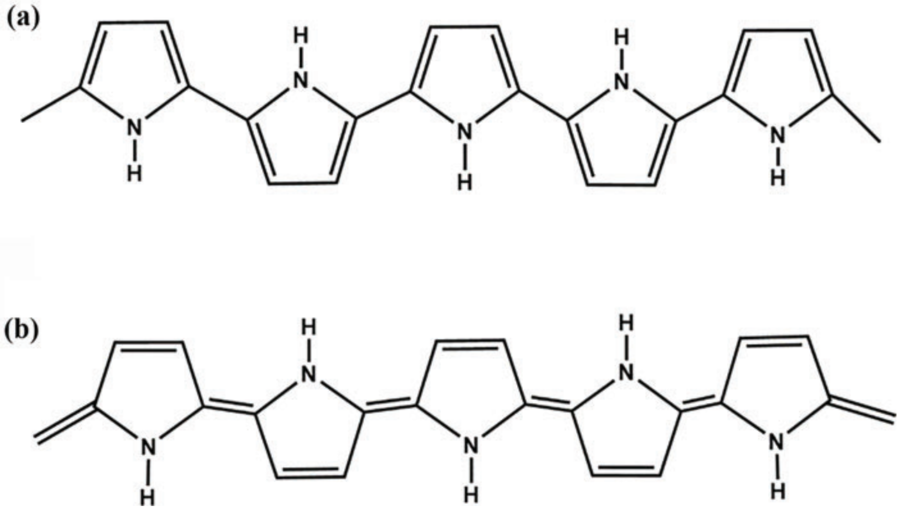

Figure 4: PPy structure in (a) aromatic and (b) quinoid forms [72]

The conductivity of PPy primarily arises from the formation of polaron and doubly charged bipolarons, which are generated when π-electrons are removed from the polymer backbone through oxidation, causing local disturbance [71,108]. PPy maintains its conductivity through the ability of reversible transformation between oxidized and reduced states. According to Eqs. (3) and (4), the reduced PPy releases a counter ion A^-, and subsequently re-oxidized by atmospheric oxygen [63,109].

$$PPy^+A^- + e^- \rightarrow PPy^0 + A^- \tag{3}$$

$$O_2 + PPy^0 + 4H^+ \rightarrow PPy^+ + 2H_2O \tag{4}$$

The conductivity of PPy depends on two key factors: (1) the number of charge carriers and (2) the mobility of these carriers. Doping modification significantly increases the density of charge carriers, thereby enhancing conductivity [110]. Additionally, many investigations have shown that the p-type doping is beneficial to improving the corrosion resistance of PPy coatings [72,111]. Therefore, exploring preeminent dopants and befitting doping processes for PPy coatings remain significant challenges for effectively protecting BPs in practical application.

Similar to PANI coatings, PPy coatings can be modified by doping with inorganic acids, organic anions, inorganics and noble metal particles [112–116]. Wang et al. optimized H_2SO_4-doped PPy coating using a Taguchi DOE method, demonstrating partial protection for the 316L SS substrate [113]. To avoid the de-doping issues associated with small anions, larger organic anions have attracted more attentions. Jiang et al. compared the use of large CSA molecules with small acid SO_4^{2-} molecules as dophants for PPy coatings [63]. It found that the CSA doping offered multiple benefits for improving the barrier quality: (1) the large spatial structured sulfonate groups were difficult to exchange with corrosive anions such as SO_4^{2-}, OH^-, resulting in repelling these anions in the solutions; (2) the released SO_4^{2-} formed insoluble complexes with Fe elements in stainless steel, maintaining the passivated state of 304 SS. Therefore, in comparison with PPy coatings doped with small acid anions (SO_4^{2-}), including PPy-SO_4^{2-} or PPy/PPy-SO_4^{2-}coatings, the PPy-CSA coating presented improved ICR and corrosion resistance during prolonged exposure time.

Doping inorganics into PPy-based composite coatings is also an effective strategy to improve their performance [116–120]. Sati et al. demonstrated that the metal oxide nanocomposites (Al_2O_3, ZnO, TiO_2, CeO_2, and SnO_2) can reduce porosity and improve the morphology of PPy coatings. The protective efficacy of these composite coatings follows the order of PPy-Al_2O_3 > PPy-ZnO > PPy-TiO_2 > PPy-CeO_2 > PPy-SnO_2 [114]. Jiang et al. investigated the use of GO as a dopant to create PPY/GO composite coatings. The introduced GO improved the adhesion of the coatings to be more uniform and compact, significantly enhancing the ability to block corrosive agents. In addition, the GO with the PPy matrix maintains the anodic protection during immersion and forms stable passive film at coating/304 SS interface. As a result, the PPy-GO composite coating effectively improves the long-term corrosion resistance of 304 SS BPs [115]. However, non-conducting inorganics have limited effects on conductivity, a critical requirement for BPs [116].

To address this, conductive dopants with strong chemical stability and good dispersion properties, such as noble metal particles, carbon-based materials and transition metal compounds, have drawn significant attention for improving both the corrosion resistance and conductivity of PPy coatings [66]. Au and Ag are the most commonly used noble metal particles for modifying PPy coatings [117–120]. Studies by Jia et al. and Ji et al. have demonstrated that the conductivity of PPy coating is profoundly improved by incorporating Ag into PPy [118,119]. Liu et al. prepared a PPy/Au composite coating on Ti-based substrate, utilizing oriented Au nanoparticles (AuNPs). This composite leveraged the highly conductive nature of AuNPs along with the physical protection provided by PPy, resulting in a coating with low ICR (initial: ~ 0.9 mΩ cm^2 at 140 N cm^{-2}; after 1000 h: 1.5 mΩ cm^2 at 140 N cm^{-2}) and excellent corrosion resistance (0.17 μA cm^{-2}) in simulated PEM water electrolysis environments [120]. Ashassi-Sorkhabi et al. also confirmed that PPy/Au nanocomposite coatings exhibit superior corrosion resistance compared to PPy alone [117]. However, the high cost and limited availability of noble metals pose significant challenges for their largescale application. To overcome this issue, exploiting and fabricating novel dopants with high conductivity and low cost are quite urgent and necessary.

Carbon materials including graphene, CNTs, carbon nanofibers and carbon powder have attracted intense interests due to their high surface area, excellent electrical conductivity, chemical stability and affordability [121]. These properties make carbon materials ideal dopants for modifying PPy coatings [122–126]. Liu et al. deposited a PPy/G composite coating on 304 SS by CV method. This composite not only demonstrated great corrosion resistance, but also retained excellent conductivity of graphene. Hence, it achieved significantly lower corrosion current density i_{corr} (9.25 μA cm^{-2})

and ICR (19 mΩ. cm^{-2}), compared with pure PPy coating (i_{corr}: 28.1 μA cm^{-2}; ICR: 41 mΩ cm^{-2}) [123]. Yilmaz et al. successfully deposited PPy/TBPB-MWCNT (TBPB-MWCNT: Tetra Butyl Phosphonium Bromide doped Multi-Walled Carbon Nanotube) composite coating on 430 SS, providing effective substrate protection [124]. Chen et al. investigated the complementary properties of PPy and CNTs by developing PPy/CNT composites, highlighting the anionic doping role of CNTs within the PPy matrix to leverage their remarkable conductivity [125]. In another study, Chen et al. utilized the polydopamine (PDA) functionalized carbon powder (C-PDA) as dopants to prepare PPy/C-PDA composite coating. The incorporating carbon powders enabled the PPy coating to self-repair under aggressive attack, ensuring excellent protection and adhesion strength with low ICR in simulated PEMFC environments [126].

In addition to carbon materials, transition metal compounds are increasingly recognized as viable dopants due to their high conductivity, chemical stability and cost-effectiveness [127]. Among them, TiN, Ti_4O_7, $Ti_3C_2T_x$ MXene are representative examples utilized in PPy coatings [127–129]. Ding et al. deposited PPy/TiN composite coatings by constant potential polymerization. As expected, the conductive TiN particles serve as “conductive bridges” within the PPy matrix, enhancing coating density and reducing both ICR and i_{corr} [127]. Tan et al. successfully prepared a PPy composite coating doped with modified Ti_4O_7 particles on 316L SS by electrochemical deposition. The Ti_4O_7 particles formed chemical bond with PPy by electrostatic interaction, improving conductivity and hydrophobicity, which proved effective in simulated cathodic PEMFCs environments [128]. Bian et al. directly doped the conductive $Ti_3C_2T_x$ MXene into the PPy matrix through one step electrodeposition, resulting in a PPy/MXene composite coating on 304 SS BPs. This MXene addition brought lots of benefits, such as improved barrier effect, anodic protection, enhanced adhesion and conductivity. Therefore, the PPy/MXene coating exhibited superior long-term stability and a significantly lower ICR (55.24 mΩ cm^2) in simulated PEMFCs environments, compared to pure PPy coating (108.16 mΩ·cm^2) [129].

Despite the promise of ICPs coatings like PANI and PPy for modifying metallic BPs in PEMFCs, several challenges remain: (1) ICP coatings typically suffer from poor adhesion to BP substrates due to the lack of polar functional groups [65,78], leading to detachment during the assembly process of PEMFC and reduce the long-term durability; (2) most studies on ICP coatings are carried out in simulated PEMFC environments. To ensure their practical viability, more investigations is needed to evaluate their performance under complex and demanding conditions of actual fuel cell assemblies [66].

4.2 NICPs Coatings

In addition to intrinsic conductive ICP coatings, the NICP coatings also have attracted significant attention from materials scientists and engineers [66]. By incorporating conducting fillers, NICP coatings can achieve both high conductivity and excellent corrosion resistance in PEMFC environments [130]. In this regard, conducting fillers plays a critical role in NICP coatings and should meet several key requirements: (1) high chemical stability to ensure durability under harsh environments; (2) ability to establish conductive pathways, which necessitates fillers with structures of at least one large dimension in three-dimensional space; (3) strong compatibility with the polymer matrix for tight bonding and homogeneous dispersion. Given these criteria, one-dimensional nanowires and two-dimensional nanosheets have become the ideal fillers for NICP coatings [66,130–134]. These nanostructured fillers, showing typical feature of large surface areas, smooth surfaces, and favorable aspect ratios, extent the diffusional path of corrosive agents, thereby enhancing the coating’s protective performance [130].

Fu et al. investigated an Ag-polytetrafluoroethylene (PTFE) composite coating electrodeposited on 316L SS. The Ag-PTFE composite coating exhibited strong hydrophobic, high interfacial conductivity and good corrosion resistance, owing to the conductivity of Ag and the barrier effect of PTFE [131]. Bedel et al. reported that the introduction of Ag nanowires into the poly (epoxy) matrix significantly reduced surface resistivity to as low as 13 mΩ sq^{-1} [132]. Chu et al. prepared epoxy/PANI nanowire composite coatings and observed improved water repellency and adhesion. The optimized composite coating containing 2 wt.% PANI nanowires exhibited excellent corrosion resistance in a severe 12 wt.% NaCl solution at 95°C [133]. Similarly, Show et al. developed CNT/PTFE composite coating on 304 SS BPs, noting a 1.6 times increase in fuel cell output power for 75 wt.% CNT coating due to its high conductivity (12 S cm^{-1}) and enhanced corrosion resistance [134,135].

In terms of two-dimensional materials, Yang et al. designed a $Ti_3C_2T_x$ MXene (T)-carbon black (C)-acrylic epoxy (AE) composite coating for 304 SS BP in PEMFCs, achieving low ICR (15.5 mΩ cm^2 under 140 N cm^{-2}) and superior long-term stability. Owing to the excellent electrical conductivity of $Ti_3C_2T_x$ MXene nanosheets and carbon black, along with the physical barrier and the cathodic protection provided by the stacked $Ti_3C_2T_x$ MXene nanosheets, the T-C-AE coating resulted in minimal metal ion dissolution and a low corrosion current density (6.73 μA cm^{-2}) [136]. Husby et al. designed a carbon-polymer composite coating with 45 vol% graphite flakes, 5 vol% carbon black and 50 vol% epoxy binder for 316L SS substrates. The conductive pathways created by the graphite resulted in a low ICR of 9.8 mΩ cm^2 under 125 N cm^{-2} [137].

Although NICP coatings with conducting fillers have demonstrated excellent conductivity and corrosion resistance, their widespread application is still limited by challenges in maintaining continuous conductive paths across the coating during largescale production [50]. In addition, achieving high conductivity often requires thinner coatings or high concentration of conducting fillers, which can lead to issues like agglomeration, high cost, reduced stability and adhesion [66]. Therefore, further research is necessary to explore advanced conducting fillers and optimize preparation processes to overcome these limitations.

4.3 Double-Layer or Multi-Layer CP Coatings

To date, numerous of CP coatings have been developed using various fabrication methods, each offering distinct advantages and limitations. In order to leverage performance by combining the advantages of different coatings, the strategy of designing double-layer or multi-layer coatings have proven extremely workable and effective. For the CPs coatings on metallic BPs, the basic principle are as follows [138–140]: (1) the inner layer should adhere well to both the metallic BP substrate and the outer layer, to avoid coating exfoliation and ICR increasing; (2) the outer layer should possess excellent corrosion resistance to corrosive media; (3) the thickness should be controllable to balance corrosion resistance and electrical conductivity.

For instance, Jiang et al. designed a PPy-GO/PPy-CSA double-layer coating on 304 SS BPs by electrodeposition. The inner PPy-GO layer strengthened the adhesion and extended the diffusion path for the corrosive species, while the outer PPy-CSA layer provided anodic protection by impeding ion exchange and preserved conductivity [138]. This design achieved superior anti-corrosion performance in simulated chloride-rich PEMFC environments. Similarly, Pan et al. designed a PPy/PANI double-layer coating on Cu BPs through electrodeposition. It is revealed that the bi-layered structure with different ion-permselective nature served as an effective physical barrier against the inward penetration of corrosive species [139]. Gao et al. further advanced this approach with a PANI-Au/Au double-layer coating on 316 SS BPs, where the inter PANI layer provided strong corrosion resistance and the outer AuNPs layer further improved both the corrosion resistance and conductivity, meeting DOE targets for ICR [140].

Although double-layer and multi-layer CP coatings can effectively enhance the corrosion resistance, they introduce certain challenges. The increased interface number may raise electrical resistance and the risk of exfoliation. In addition, the multi-layer preparation process is more complex than single-layer coatings, which could impact scalability and production costs.

Finally, Table 4 provides a comprehensive summary of the synthesis methods for CP coatings on metallic bipolar plates, the simulated PEMFC experimental processes and the corrosion resistance and conductivity performance results discussed in the preceding sections. This overview offers a clearer and more intuitive understanding of the current research progress on CP coatings for metallic bipolar plates.

Table 4: Summary of synthesis methods, experimental conditions and performance metrics for CP coatings on metallic bipolar plates [49,63,91,98,99,102–104,120,123,126,139]

Substrates	Coatings	Synthesis methods	Electrolytes	i (μA cm^{-2})	ICR (mΩ cm^2)	Ref.
304SS	PPy-CSA	Galvanostatically (camphorsulfonic acid)	0.1 M H_2SO_4	1.87×10^{-9}	5.5	[63]
316L SS	PANI	Galvanostatic technique (H_2SO_4)	+ 0.5 M H_2SO_4 + 2 ppmHF, 80°C	9.3×10^{-8}	/	[49]
316L SS	PANI	Cyclic voltammetry (H_2SO_4)	1 M H_2SO_4 + 2 ppm HF, 70°C	$<1.62 \times 10^{-6}$	<10	[91]
303SS	PANI/Zn-Pr	PANI/Zn-Pr was coated by spray gun and were cured at 70°C for 1.0 h	1.0 M H_2SO_4, 25°C	1.5×10^{-7}	/	[98]
304SS	PANI-TiN	Cyclic voltammetry	1.0 M H_2SO_4, 80°C	5×10^{-7}	32	[99]
316L SS	PANI/CNTs	Direct current electrodeposited (H_2SO_4)	0.1 M H_2SO_4 + 2 ppm HF, 80°C	1.9×10^{-6}	/	[102]
Al	PPy/CNTs	Chemical coating	0.1 M H_2SO_4, 25°C	1.8×10^{-7}	/	[103]
316L SS	PANI/G	*In-situ* polymerization	/	8.8×10^{-7}	10	[104]
Ti	PPY/Au	constant potential	0.5 M H_2SO_4 + 2 ppm HF	1.7×10^{-7}	1.5	[120]
304SS	PPy/G	Cyclic voltammetry (SDS)	0.5 M H_2SO_4 + 2 ppm HF, 25°C	9.25×10^{-6}	19	[123]
304SS	PPy/C-PDA	Cyclic voltammetry (oxalic)	0.1 M H_2SO_4	9.25×10^{-6}	17	[126]
Cu	PPy/PANI	Cyclic voltammetry (oxalic) and Galvanostatic technique (H_2SO_4)	0.2 M H_2SO_4 + 0.1 M HCl +3 ppm HF, 25°C	1.62×10^{-6}	/	[139]

5 Life Cycle Assessment and Environmental Impact

To facilitate industrial applications, the economic and environmental sustainability of CP coatings has become increasingly important. In addition to performance improvements, the materials used for protective coatings must be cost-effective, scalable, and environmentally friendly to support the widespread adoption of fuel cell technology. CP coatings, particularly PANI and PPy, show significant potential in terms of both economic efficiency and environmental stability, and are widely recognized as conductive polymers in industrial applications [141]. However, research in this area remains limited, highlighting the need for further exploration.

Shekhar et al. [142] conducted a life cycle assessment (LCA) of PANI-GO coatings using the IMPACT 2002+ methodology, systematically evaluating the materials and processes involved in the production of PANI-GO nanocomposites. Their study revealed that the production of PANI-GO nanocomposites is energy-intensive, primarily due to the energy consumption of equipment and machinery used during synthesis, with non-renewable energy sources contributing the most significant environmental impacts. The choice of materials also significantly affects categories such as aquatic and terrestrial acidification, non-carcinogenic and carcinogenic releases, primarily due to the ecological toxicity of chemicals (e.g., hydrochloric acid, sulfuric acid, and potassium permanganate) and waste emissions (gaseous and liquid) generated during synthesis. Furthermore, the study emphasized that scaling up manufacturing processes could reduce the relative impact of emissions and energy consumption. Optimizing production techniques and incorporating renewable energy sources could substantially mitigate the overall environmental burden. Although high energy usage and chemical consumption at the laboratory scale negatively impact LCA results, these issues may not necessarily indicate excessive resource depletion. The findings provide valuable insights for the scale-up of manufacturing processes and for identifying environmental hotspots for future optimization efforts.

6 Conclusions and Future Insights

Hydrogen energy is increasingly recognized as a promising solution to address environmental pollution, resource scarcity, and energy shortages. Advancing PEMFCs is pivotal to enabling the largescale deployment of clean hydrogen energy within the global energy transition. As a key component, selecting suitable bipolar plates (BPs) and designing advanced protective coatings are essential steps in bringing PEMFCs closer to commercial reality. This review summarizes advancements in various types of BPs and protective coatings for metallic BPs in PEMFCs, with a particular focus on CP coatings.

The primary developmental directions for BPs that show potential to meet DOE targets include graphite BPs, metallic BPs, and composite BPs. Among these, metallic BPs offer broader advantages in terms of supporting membrane-electrode assembly and enabling largescale manufacturing due to their strong mechanical properties, cost-effectiveness, low gas permeability, and good electrical and thermal conductivities. In contrast, graphite BPs are constrained by low mechanical strength and poor machinability, while composite BPs face challenges related to complex production processes and relatively high costs. Surface modification treatments are, however, necessary for metallic BPs to address limitations such as increased ICR caused by passive film formation and the durability of metallic materials in harsh environments.

CPs have attracted considerable attention as promising protective barriers for BPs due to their chemical inertness and conductive properties. Research efforts have focused on optimizing CP coatings to reduce ICR, improve corrosion resistance and enhance cost-efficiency. Doping modifications have proven to be an effective strategy for enhancing these properties, with advancements in ICPs, NICPs and multi-layer CPs coatings. Dopants such as inorganic acids, organic anions and inorganic fillers have significantly improved performance by increasing conductivity and providing robust corrosion protection. Despite these advancements, achieving large-scale commercial applications for CP coatings requires addressing key challenges, including:

(1) Most current research on CP coatings has been conducted at the laboratory scale. Long-term performance and stability under actual PEMFC operating conditions remain critical factors for largescale commercial applications. Future studies should focus on evaluating CP coatings in real-world environments to ensure their reliability and durability.

(2) Transitioning CP coatings from laboratory research to industrial applications requires scalable and cost-effective manufacturing techniques. Establishing standardized production protocols and evaluation methods will ensure consistent quality and enable broader adoption in the market.

(3) Developing novel dopants and CP formulations with improved conductivity, corrosion resistance, and mechanical properties is a promising area of research. Introducing polar functional groups

into intrinsically conducting polymers (ICPs) could address adhesion issues and enhance long-term stability.

(4) Combining CPs with other materials, such as carbon-based nanomaterials or ceramic particles, offers opportunities to create multifunctional coatings. These hybrid coatings could simultaneously enhance conductivity, corrosion resistance, and thermal stability, addressing multiple performance metrics in a single system.

(5) Supportive government policies and industry initiatives are crucial to accelerating the development of the hydrogen energy transition. These efforts should focus on developing production equipment, securing material supply chains and, establishing standardized evaluation methods, fostering innovation and reducing barriers to commercialization.

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