

Preparation and Application Progress of Graphene/Polyaniline Composites

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Abstract

Graphene has excellent thermal conductivity, electrical conductivity, mechanical properties and large specific surface area. The conductive polymer polyaniline has low cost, simple synthesis and excellent electrochemical performance. This paper briefly introduces the preparation method of graphene/polyaniline composites. The applications of graphene/polyaniline composites in the fields of supercapacitors, sensors, thermoelectric materials, ion adsorption and bipolar plates are introduced in detail. Finally, the existing problems and development directions of graphene/polyaniline composites in the field of bipolar plates are prospected.

Keywords

Graphene; Polyaniline; Composite Material; Preparation Method; Apply.

1. Introduction

Graphene and polyaniline have been the focus of research because of their excellent physical and chemical properties. However, the strong π - π interaction between graphene sheets makes it easy to attract each other and agglomeration occurs, which limits the processability^[1-3]. Polyaniline has poor mechanical properties and is prone to oxidative degradation in the air, resulting in poor long-term stability^[4-6]. Therefore, the composite of graphene and polyaniline can make up for the shortcomings of both to a certain extent, give full play to the synergistic effect between graphene and polyaniline, prevent the agglomeration between graphene sheets, and improve the mechanical properties and chemical stability of polyaniline, so that the composite material has good mechanical and electrochemical properties^[7-10].

2. Preparation Method of Graphene/Polyaniline Composite Materials

There are many methods for preparing graphene/polyaniline composites, including solution blending^[11-13], in-situ polymerization, electrochemical polymerization, interfacial polymerization^[14-16], hydrothermal method^[17-20] and layer by layer autonomous assembly^[21-23]. Two chemical oxidation methods, in situ polymerization and electrochemical polymerization, are described in detail.

2.1 In-situ Polymerization

In-situ polymerization method refers to the addition of aniline monomer into the dispersion solution of graphene. Aniline is adsorbed on the surface of the graphene sheet through hydrogen bonding and electrostatic interaction, and then the addition of oxidant triggers the polymerization of aniline to obtain graphene/polyaniline composite.

Wang et al.^[24] Firstly added aniline monomer into graphene/hydrochloric acid suspension to make aniline uniformly adsorbed on the surface of graphene by electrostatic attraction, and then added

ammonium persulfate drop by drop to polymerize aniline into polyaniline molecules. Through the π - π conjugation interaction between polyaniline and graphene, polyaniline molecules are initially oriented on the surface of graphene. In the subsequent process of solution treatment, the order degree of molecular arrangement of polyaniline was further improved. The uniform dispersion of graphene provides a larger contact area for graphene and polyaniline, enhancing the π - π -conjugated interaction between them, thus promoting the formation of a more expansive molecular conformation of polyaniline molecular chains (Figure 1a). These ordered regions improve the Seebeck coefficient and TE properties of the composites. Lin et al. [25] obtained p-phenylenediamine modified graphene containing amino-aniline groups by grafting the epoxide group on the edge of graphene. The modified graphene is polymerized with aniline monomers to form a composite consisting of a very unique semi-interpenetrating network of chemically bonded two-dimensional structures (Figure 1b). By enhancing the degree of conjugate crosslinking between graphene and polyaniline, it not only provides an additional route for the smooth transport of charge carriers, but also significantly reduces the amount of graphene required to a few percentage points (≤ 3 wt %), making it possible to improve the efficiency and performance of graphene as a modified additive for the development of TE polymers.

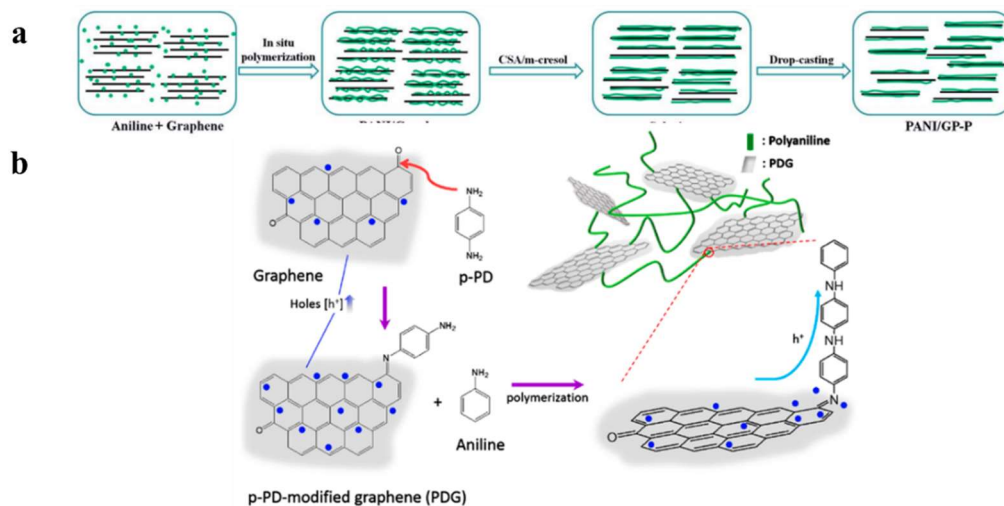


Figure 1. (a) Schematic diagram of the synthesis process of PANI/GP-P composite film [24]; (b) Schematic diagram of S-IPNs constructed by chemically bonded PDG and linear polyaniline [25]

2.2 Electrochemical Polymerization Method

Electrochemical polymerization method refers to the aniline monomer in the electrolyte under the action of electric field force, oxidation polymerization reaction on the anode to generate a film or powder attached to the surface of the anode substrate, which can control the degree of oxidation of polyaniline and the thickness of the film layer by adjusting the potential and electricity during aniline polymerization. Electrochemical polyaniline can be deposited directly on the substrate without solving the problem of film formation caused by poor solubility of polyaniline.

Altinci et al. [26] deposited graphene oxide-polyaniline (GO-PANI) composite films on platinum-iridium (Pt-Ir) electrodes by electrochemical polymerization of aniline at constant potential. The electrochemical degradation efficiency of active blue 4 and acid red 97 were 9.4% and 15.2% respectively in 0.5 M H₂SO₄ electrolyte solution within 120 min reaction time. The graphene oxide-polyaniline deposited Pt-Ir electrode had good cycling stability and reutilization. It can be used for anodizing organic matter. Pereira1 et al. [27] in 0.1 M H₂SO₄ electrolyte and 0.1 M aniline monomers solution, graphene oxide (GO) and polyaniline (PANI) were electropolymerized during the graphite stripping process, and the polyaniline was distributed between the graphene oxide multilayer films in the shape of nanoneedles. At a scanning rate of 1 mVs⁻¹, the specific capacitance of GO/PANI composite is 117.440 Fg⁻¹, while that of GO is 1.243 Fg⁻¹.

3. Progress in the Application of Graphene/Polyaniline Composites

3.1 Supercapacitor

Energy storage systems such as batteries, fuel cells, electrochemical capacitors and solar cells are currently the focus of research. Every portable electronic device requires a high power density for a long time. Due to their electrochemical performance compared to other energy storage devices, supercapacitors (SCs) have proven that they can do this. Fu et al. [28] prepared Nafion-doped polyaniline/graphene oxide composites as electrode materials by in-situ polymerization method, and designed a flexible supercapacitor based on Nafion film (Figure 2). After Nafion doping, the utilization and electrochemical surface area of polyaniline are improved, and the specific capacitance of capacitors under horizontal and bending conditions (1 A/g) is 309 and 300 F/g, respectively, which is the maximum value reported for Nafion membrane-based supercapacitors. After 1000 cycles, the capacitance retention of the supercapacitor using the Nafion membrane is 87.2%, which is higher than the 83.0% of the H₂SO₄ electrolyte.

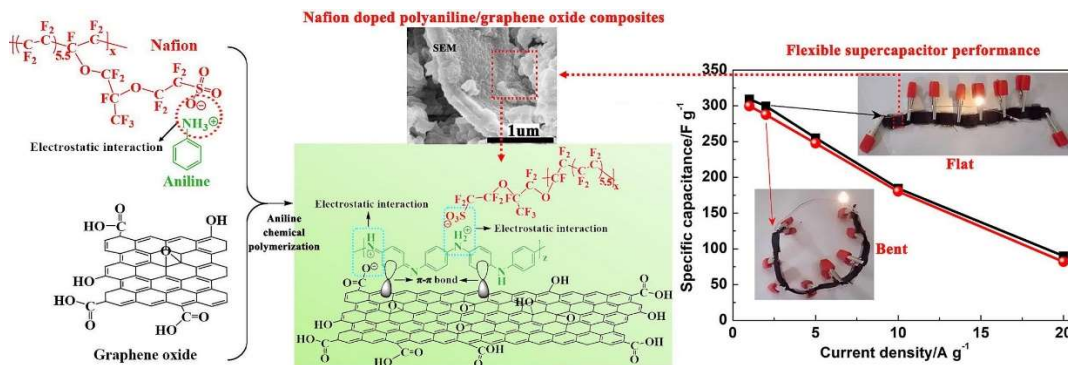


Figure 2. Preparation process of Nafion-doped polyaniline/graphene oxide composites, SEM image of the composites and specific capacitance of flexible supercapacitors at 10 A/g [28]

Xu et al. [29] also used in-situ polymerization method to prepare terpolymer materials composed of polyaniline, porous carbon C800 and low oxide graphene LGE, and used a simple stamping method to manufacture flexible paper-based micro-supercapacitors. Under the synergistic action of porous carbon C800 and low graphene oxide LGE, the conductivity and capacitance retention of polyaniline composites are significantly improved. The prepared composite material has a high surface capacitance of 162 mF cm⁻² (Figure 3a), an energy density of 24.9 μWh cm⁻² (Figure 3b), 90% capacitance over thousands of charge and discharge cycles, and excellent bending stability even at large angles.

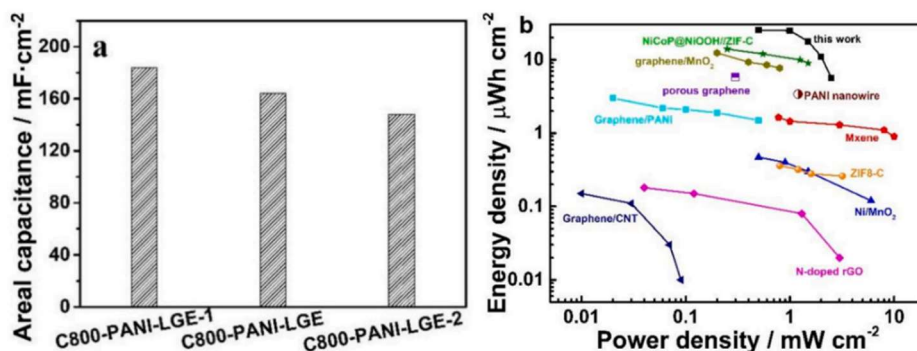


Figure 3. (a) Surface capacitance of MSCs based on C800-PANI-LGE, C800-PANI-LGE-1 and C800-PANI-LGE-2 at a scanning rate of 10 mV•s⁻¹; (b) This study is based on the Ragone diagram of C800-PANI-LGE's artificial MSC and other reported MSC systems [29]

Patil et al. [30] reported a composite material consisting of α -manganese dioxide (α -MnO₂) coated polyaniline (PANI) and reduced graphene oxide (rGO) for supercapacitors. In situ polymerization of polyaniline in the presence of metal oxides, high quality composites were obtained. The specific capacitance of α -MnO₂/PANI composites was improved by adding carbon-based materials such as rGO. Reduced graphene oxide in α -MnO₂/PANI composites is extremely important for obtaining high-quality composites. In addition, the in-situ polymer coating on the metal oxide greatly improves the film forming properties of the material and enhances the machinability of the material in device manufacturing. The specific capacitance of α -MnO₂-coated PANI/rGO nanocomposites is 261 F g⁻¹ at 5 mV s⁻¹ scanning rate, and the capacitance retention of t is 75% after 2000 cycles at 5 A g⁻¹, the specific energy is 11 W h kg⁻¹, and the specific power is 1250 W kg⁻¹(Figure 4a, b).

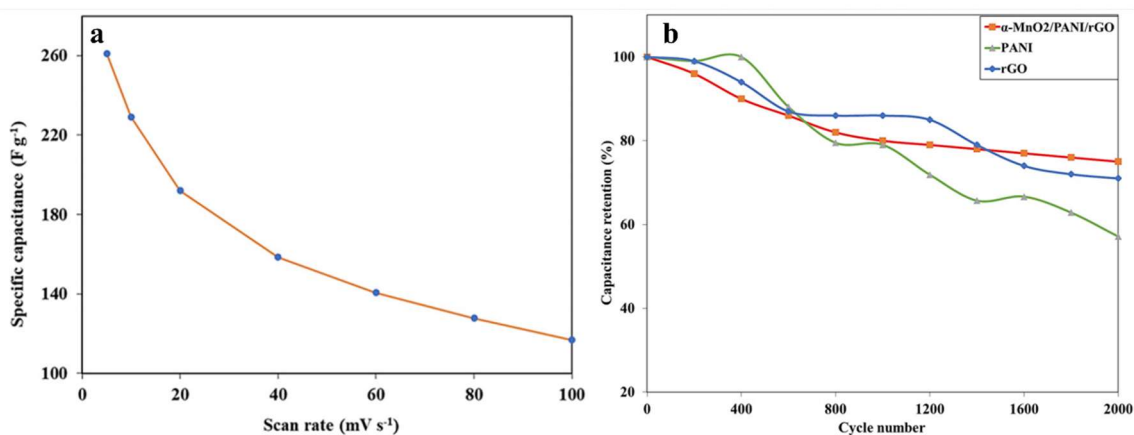


Figure 4. (a) The relationship between scanning rate and specific capacitance of α -MnO₂/PANI/rGO; (b) Cyclic stability of polyaniline, graphene oxide and α -MnO₂/ polyaniline/graphene oxide nanocomposites [30]

3.2 Sensor

Due to the growing concern for human health and environmental protection, the development of sensitive devices for the detection of toxic chemicals has recently attracted considerable interest. Ammonia (NH₃) is one of these toxic chemicals and has applications in many industries. Therefore, it is not far-fetched to think that the development of highly sensitive and cost-effective NH₃ monitoring sensing devices is critical. Tabar et al. [31] proposed a novel ammonia (NH₃) chemical sensor with ultra-high response, high selectivity, fast response and long-term stability by using a polyaniline/copper oxide nanoparticle detection layer supported by three-dimensional nitrogen-doped graphene skeleton (PANI/CuO@3D-NGF) nanocomposites. Polyaniline /CuO@3D-NGF nanocomposites were prepared by in-situ polymerization of polyaniline on CuO@3D-NGF with high surface area. Morphological and structural analyses show that the ultra-thin three-dimensional interconnected graphene substrate maximizes the surface area. The results show that the nano-CUO provides an active adsorption site for free NH₃ molecules. The PANI/CuO@3D-NGF nanocomposite gas sensor has a response rate of 930% to 100 ppm NH₃, a low detection limit of 50 ppb at room temperature, and an average response time of 30 s(Figure 5a). The excellent sensing properties of polyaniline /CuO@3D-NGF nanocomposites are mainly attributed to the three-dimensional interconnected porous structure, the remarkable enhancement of CuO@3D-NGF carriers and the modification of the intermolecular π - π interaction.

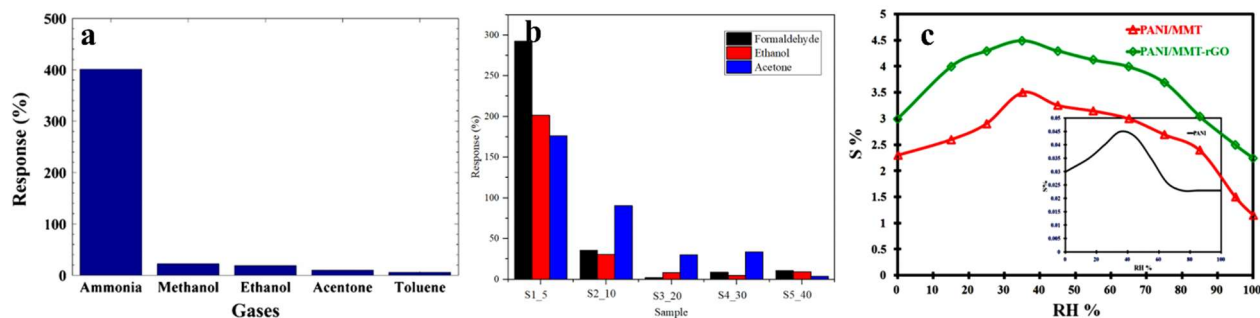


Figure 5. (a) Comparison of selectivity of PANI/CuO@3D-NGF nanocomposite gas sensor when exposed to 40 ppm of various VOC vapors at $25\pm 2^\circ\text{C}$ and $56\pm 2\%$ RH^[31]; (b) Selective response of IDE sensors to 1000ppm formaldehyde, ethanol and acetone in the room^[32]; (c) Sensor response to HCN (2ppm) starting from different RH values^[33]

The performance of conventional metal oxide sensors (MOX) is largely dependent on their high operating temperatures, and many researchers have tried to solve this problem by exploring composite materials. Hussein et al.^[32] synthesized a ternary hybrid material consisting of metal oxide (Fe_3O_4), polymer (polyaniline) and carbon-based material (reduced graphene oxide) by in-situ polymerization method. Hybrid nanocomposites were formed through strong interaction between the components. When the Fe_3O_4 -PANI-RGO mass fraction is 10 wt%, the response and recovery time to acetone concentrations of 1, 10, and 100 ppm are < 32 s, with the highest sensing response at room temperature (Figure 5b). Singh et al.^[33] synthesized polyaniline montmorillonite reduced graphene oxide polymer nanocomposites by chemical oxidation polymerization of aniline with ammonium persulfate and sulfuric acid for the detection of hydrogen cyanide (HCN) vapor. When the sensor is exposed to different gases acetone, ammonia, benzene, hydrogen cyanide and xylene, the sensor material detects HCN gas and gives the highest sensing response (Figure 5c). At 2 ppm hydrogen cyanide (HCN) gas, the sensor achieved a 4.56% sensing response to the PANI/MMT-rGO. The sensing material polyaniline alone has a low sensing response of 0.05%. With the addition of MMT and rGO, the sensing response of the sensing material increases, and the sensitivity of PANI/MMT and PANI/ MMT-RGO are 0.89 ppm^{-1} and 1.1174 ppm^{-1} , respectively. The improved sensor sensitivity may be due to the increased surface area provided by MMT and rGO, providing more binding sites for HCN gas. In both sensors, the sensing response increases as the exposed gas concentration increases, with automatic recovery within 21 s (PANI/MMT) and 25 s (PANI/ MMT-RGO). Sensor performance declined after 6 and 8 months.

3.3 Thermoelectric Materials

Thermoelectric materials have always been an object of interest for energy demand because of their potential for heat capture and their potential for conversion into electrical energy, with about 60% of the total energy wasted in the form of thermal energy, thermoelectric materials (TEMs) can avoid this heat waste and help reduce the energy crisis by harnessing their ability to collect and convert waste heat into electrical energy. Jorge et al.^[34] prepared hybrid nanocomposites by mixing graphene oxide (GO) and nano-needle (n-ZnO) and nano-flower (f-ZnO) nanoparticles into polyaniline matrix with m-cresol solution. The ternary hybrid nanocomposites prepared by graphene oxide/zinc oxide have better thermal stability than polyaniline/zinc oxide. In terms of thermoelectric properties, polyaniline with better electrical conductivity has been successfully synthesized, and the dopants and graphene oxide/zinc oxide nanoparticles used provide greater electrical conductivity for the final nanocomposites. The Seebeck coefficient of PANI ZnO nanocomposites is the highest, and the Seebeck coefficient is the highest when PANI/n-ZnO is 2.5%. PANI/GO/n-ZnO has the highest thermoelectric factor, and its TPF value is $14.79 \mu\text{W m}^{-1} \text{K}^{-2}$, which is 49.3 times that of pure polymer and 1.5 times that of PANI/n-ZnO nanocomposites (Figure 6a, b).

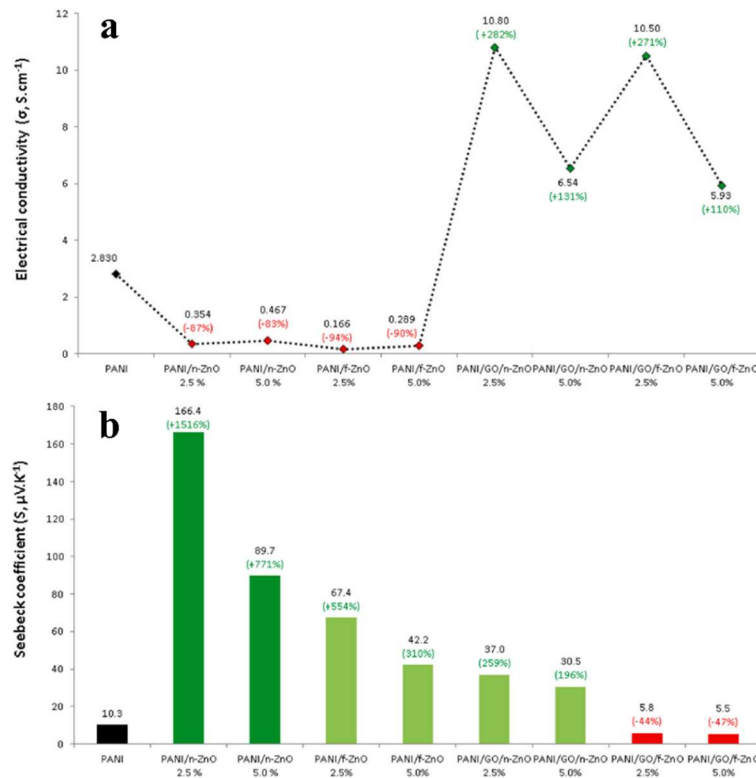


Figure 6. (a) The conductivity of PANI and PANI/n-ZnO, PANI/f-ZnO, PANI/GO/n-ZnO, PANI/GO/f-ZnO nanocomposites at 2.5% and 5.0% nanoparticle content, respectively; (b) Seebeck coefficient values of PANI/n-ZnO, PANI/f-ZnO, PANI/GO/n-ZnO and PANI/GO/f-ZnO nanocomposites with 2.5% and 5.0% nanoparticle content [34]

Ali et al. [35] used nitrogen doped and sulfur nitrogen co-doped reduced graphene oxide (rGO) to regulate the thermoelectric properties of polyaniline, and prepared doped graphene oxide and polyaniline/doped graphene oxide nanocomposites by hydrothermal method and chemical oxidation polymerization method respectively. The Seebeck coefficient, power factor and zT values of polyaniline nanocomposites containing 1 wt% sulfur nitrogen co-doped graphene oxide are -1.75 mV K^{-1} , $95 \text{ μW m}^{-1} \text{ K}^{-2}$ and 0.06, respectively. It was suggested that polyaniline/heteroatom-doped graphene oxide could be a promising candidate for N-type thermoelectric applications (Figure 7a, b, c).

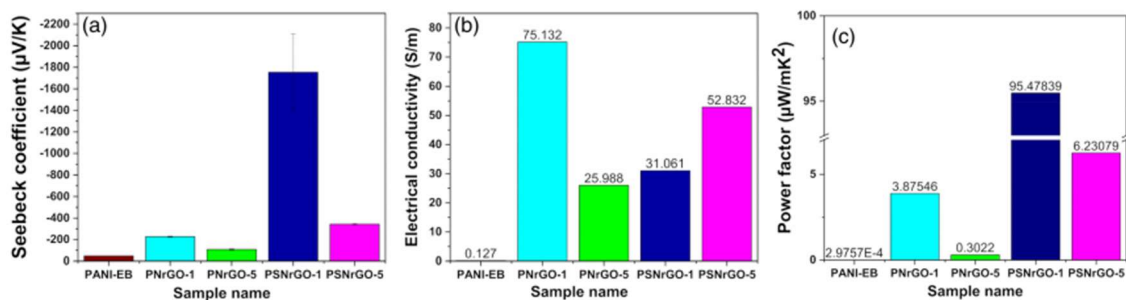


Figure 7. Different components (a) Seebeck coefficient; (b) Electrical conductivity; (c) Power factor of polyaniline and its nanocomposites [35]

3.4 Ion Adsorption

The large surface area of graphene, the high adsorption capacity of aromatic ring containing pollutants and the good complexation of heavy metals by N atoms in the molecular chain of polyaniline make graphene/polyaniline composites expected to have good adsorption properties for heavy metal Pb (II)

and aromatic ring pollutants methylene blue. It can be used as a good adsorbent in wastewater treatment where heavy metals and aromatic pollutants coexist. Xie ^[36] et al. grafted p-phenylenediamine on the surface of graphene oxide (GO), used the amino group exposed on the surface of graphene oxide as the active site of aniline in-situ REDOX polymerization, and grafted polyaniline molecular chains on the edge of graphene oxide, so that graphene and polyaniline had higher thermal stability and adsorption capacity for lead ions and organic dyes. The maximum adsorption capacity of Pb⁺ by GO-PANI composite is 1416 mg/g, which is 2.3 times of the maximum of PANI, indicating that graphene/polyaniline composite can be used as an excellent adsorption material for Pb²⁺ heavy metal ions. Kuznetsova et al. ^[37], based on reduced graphene oxide and carbon oxide nanotubes, modified by polyaniline and phenolic resin, prepared a new nanocomposite material that could effectively adsorb and remove toxic lead (II) from water by carbonizing the original aerogel. At 77 K, the porosity of the sample was estimated by nitrogen adsorption. The results show that carbonized aerogels are mainly mesoporous materials with a specific surface area of 315 m²/g. After carbonization, the small and micro pores increase, and the high pore structure of the carbonized composite is preserved. In static mode of carbonized materials, the maximum adsorption capacity of carbonized aerogel for Pb(II) is 185 mg/g at pH 6.0, the desorption rate is very low (0.3%) at pH 6.5, and the desorption rate is about 40% in strongly acidic media.

3.5 Bipolar Plate

Proton exchange membrane fuel cell is a clean energy source fueled by hydrogen, which has a broad application prospect in new energy vehicles and other fields. Bipolar plate is an important component of PEMFC, which needs to have good electrical conductivity and corrosion resistance. Stainless steel is an ideal candidate material for bipolar plates because of its good mechanical properties, good electrical conductivity, low price and easy processing. However, in the weakly acidic working medium of PEMFC, the surface of stainless steel is easy to form passivation film, which affects the working efficiency of fuel cells, so the surface treatment or coating protection of stainless steel is needed to improve the service life and chemical stability of the bipolar plate. Li et al. ^[38] firstly electrochemically polymerized the polyaniline substrate by cyclic voltammetry, and then added graphene oxide aqueous solution onto the substrate by CV electrochemical reduction to obtain rGO/PANI composite coating. rGO was uniformly covered on the surface of short rod-like PANI to fill the gaps and holes caused by PANI accumulation. Effectively prevent the corrosion material from spreading to the substrate (Figure 8a, b). The rGO/PANI double-layer composite film was soaked in 0.2 mol/L H₂SO₄ solution for 56 days, and the corrosion process of the coating could be divided into three stages: At the initial stage of immersion (0 ~ 8 d), the corrosive medium corroded the graphene, dissolved and fell off, and the surface layer of the coating gradually thinned, resulting in the open potential decreasing from 0.125 V(7 h) to 0.116 V(8 d), and the charge transfer resistance R_{ct} decreasing from 589.9 Ω (0 h) to 384.1 Ω (8 d). The R_{ct} decreases gradually and the conductivity of the coating increases. During the middle stage of soaking (9 ~ 25 d), the open circuit potential changed dramatically, and suddenly dropped to 0.109 V at 12d, and then began to rise to 0.113 V at 15 d, and then gradually dropped to 0.108 V at 25 d, which was similar to the open circuit potential value at 12 d. The R_{ct} increased from 469.5 Ω(10 d) to 504.9 Ω(21 d) and then decreased to 429.1 Ω(23 d). This process is related to the formation of passivation film between the PANI coating and the stainless steel substrate, the process of corrosion dissolution, and self-repair. PANI may react with Fe on the stainless steel surface in the presence of water and oxygen to form a metal complex on the substrate surface. PANI can improve the open circuit potential of the coating through its own REDOX reaction to achieve the purpose of self-repair. At the later stage of immersion (27 ~ 56 d), the open circuit potential of the coating slowly decreased and finally stabilized at about 0.09 V. When PANI was oxidized, the R_{ct} increased from 467.5Ω (27 d) to 634.7 Ω (56 d). The overall conductivity of the coating decreased, and the coating surface remained intact after 56 days of immersion. No stainless steel substrate is exposed, indicating that the rGO/PANI composite coating has a better protective effect on the metallic stainless steel.

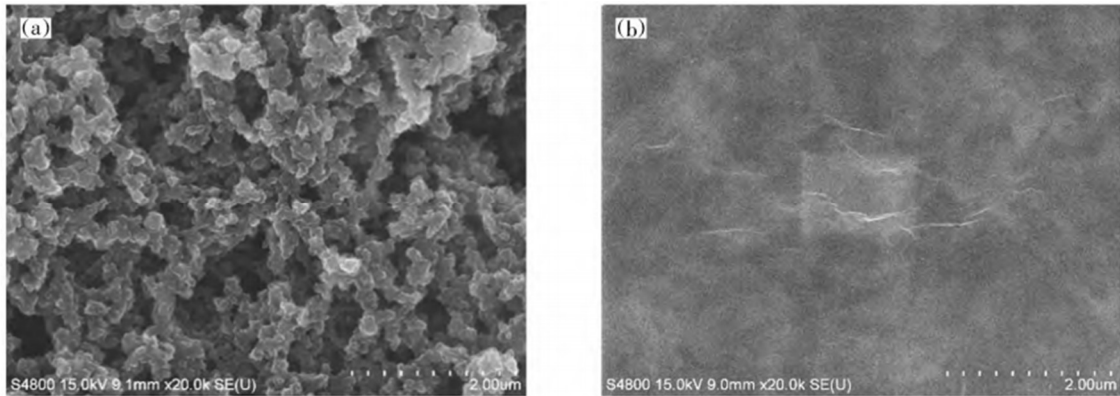


Figure 8. (a) Plane scan of pure PANI coating; (b) Plane scan of rGO/PANI composite coating [38]

Similarly, Li et al. [39] also tested the corrosion resistance of rGO/PANI composite coating under 0.2 mol/L H₂SO₄ aqueous solution and three gases. Compared with single-layer PANI coating and stainless steel bare plate, rGO/PANI composite coating exhibits higher open circuit potential, lower corrosion current density and larger potential passivation interval (Figure 9a, b, c), indicating that rGO/PANI composite coating has better corrosion resistance and stability.

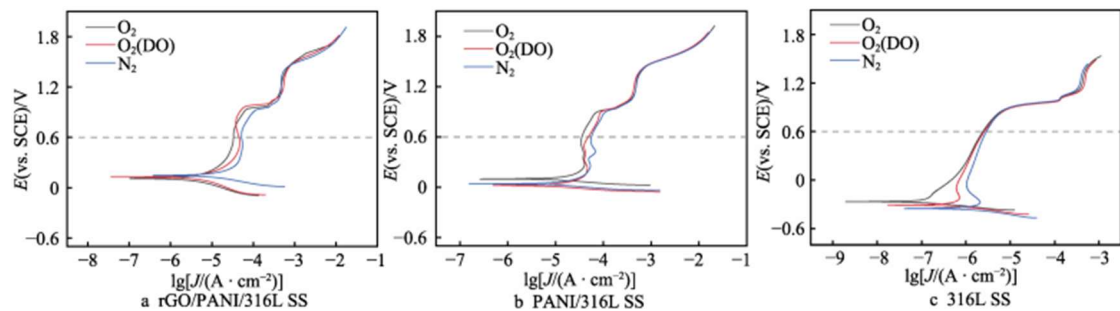


Figure 9. Potentiodynamic polarization curves of three different systems in 0.2 mol/L H₂SO₄ aqueous solution [39]

Table 1. Polarization curve data of three different systems in 0.2 mol/L H₂SO₄ aqueous solution

Coatings	Corrosive environment	$J_{corr}/(\mu A \cdot cm^{-2})$	$E(vs.SCE)/V$
rGO/PANI/316L SS	N ₂	52.5	0.71
	O ₂ (DO)	45.2	0.73
	O ₂	33.1	0.74
PANI/316L SS	N ₂	57.5	0.67
	O ₂ (DO)	48.9	0.68
	O ₂	36.3	0.69
316L SS	N ₂	2.95	1.11
	O ₂ (DO)	2.63	1.10
	O ₂	2.45	1.04

3.6 Application of Graphene/Polyaniline Composites in Other Aspects

Corrosion of metallic materials causes significant economic losses and safety threats to many facilities in the Marine environment, and functional polymer coatings are widely used to solve challenging problems in industry and are considered one of the simplest and most effective ways to prevent such adverse events. Huang et al. [40] prepared GO-PANI-PDA nanocomposites by in-situ chemical polymerization of dopamine (DA). The coating exhibits the best corrosion resistance at a 2:1 ratio of epoxy to GO-PANI-PDA. The corrosion potential of epoxy resin /GO-PANI is increased from -0.61 V to -0.51 V, and the corrosion current density is decreased from 7.05×10^{-7} A/cm² to 3.83×10^{-8} A/cm², which is an order of magnitude decrease. In the 30-day salt spray test, the coating has no obvious corrosion phenomenon, and the surface has good anti-corrosion performance. Polyaniline (PANI) has excellent REDOX properties, allowing reversible doping/dedoping and adjustable oxidation levels, making it ideal as a filler for forming conductive networks inside 3D printed resins. Graphene can achieve additional electron delocalization inside the composite by interacting with the π - π stack of polyaniline nanomaterials, acting as a filler with very high reinforcement efficiency within the polymer matrix. The synergistic effect between polyaniline nanomaterials and graphene-based materials will improve the electrical and mechanical properties of ABS-like resins. Jang et al. [41] successfully prepared ABS-like resin composite inks by adding 3.0 wt.% polyaniline nanofibers and 1.5 wt.% graphene sheets, and obtained products with high electrical conductivity and mechanical robustness with ideal shapes and different sizes through DLP(digital light processing technology) 3D printing. The wafer resistance of the 3D printed composite was reduced from 2.50×10^{15} ohm/sq (the wafer resistance of the original ABS-like resin) to 1.61×10^6 ohm/sq. In addition, compared with the original ABS resin, the tensile strength of the AP3.0G1.5 sample (3D printed composite containing 3.0 wt.% polyaniline nanofibers and 1.5 wt.% graphene sheets) was improved by 2.63 times (22.23 MPa), and the Young's modulus was increased by 1.47 times (553.8 MPa). Elongation at break is increased by 5.07 times (25.83%), tensile strength is 8.46 MPa, Young's modulus is 376.6 MPa, elongation at break is 5.09%.

4. Conclusion and Prospect

By forming a composite material of polyaniline and graphene, the agglomeration of graphene and the poor chemical stability of polyaniline can be improved. Excellent properties can be obtained by combining the two to form a composite material, which has broad and mature applications in the fields of supercapacitors, sensors, heavy metal ion adsorption, thermoelectric materials, Marine corrosion prevention, etc.

However, the current research on bipolar plates in proton exchange membrane fuel cells remains to be further explored. Combined with the performance indicators of the US Department of Energy US-DOE 2020 for bipolar plates (corrosion current density ($I_{\text{corr}} < 1 \mu\text{A} / \text{cm}^2$; Contact resistance (ICR) $< 10 \text{ m}\Omega \cdot \text{cm}^2$; Conductivity $> 100 \text{ S/cm}$), pointed out some existing problems of graphene/polyaniline composites in the field of bipolar plates, and put forward reasonable guesses and suggestions:

1) Bipolar plates have extremely high requirements for electrical conductivity, but the graphene polyaniline composite coatings currently applied to bipolar plates are based on reduced graphene oxide, and the electrical conductivity is far from meeting the requirements. Whether the reduction degree of graphene oxide can be further improved, or the graphene nanosheets can be chemically modified by grafting oxygen-containing groups on their edges to improve the dispersion of graphene sheets while retaining high electrical conductivity;

2) The surface contact resistance of the bipolar plate has a great relationship with the flatness of the coating surface, and suitable experimental parameters can be selected or some surfactants can be added to improve the surface topography of the coating when electrochemical polymerization is used. When preparing polyaniline by in-situ polymerization method, sulfonic acid doping can be used to optimize the solubility of polyaniline, and modified graphene with good electrical conductivity and dispersion can be used to prepare composite materials. Continuous centrifugal spraying or ultrasonic

spraying can be used to prepare coatings on the surface of bipolar plates. The continuous centrifugal spraying method uses a high-speed rotating drum to generate centrifugal force and shear force, so that the graphene nanosheets are parallel to the substrate and closely arranged, obtaining a flat surface topography while maximizing the "maze effect" of graphene. Ultrasonic spraying method means that the nozzle has ultrasonic effect, and under the pressure of inert gas, the coating can be atomized to the nanometer level and evenly distributed on the substrate surface, which can effectively prevent the agglomeration of graphene and obtain a good surface topography;

3) Although the current research on the performance of the bipolar plate coating mainly focuses on its corrosion resistance, there is still a certain distance from the hard index of the corrosion current density of the bipolar plate set by the United States Department of Energy. Whether the structure of the composite material can be effectively controlled and the multi-layer structure can be realized to improve the corrosion resistance of the coating. Or through the method of coating post-treatment, the coating is inhaled into the resin with small molecular weight, and the sacrifice of surface contact resistance is controlled in a reasonable range, and the corrosion resistance of the coating is improved as much as possible.

Therefore, it is still difficult for graphene/polyaniline composites to make new breakthroughs in the field of proton exchange membrane fuel cell bipolar plates, and future research must continue to ensure that graphene/polyaniline composites with new structures and excellent properties are better applied to people's actual lives.

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