

A Review on the Synthesis Methods, Properties, and Applications of Polyaniline-Based Electrochromic Materials

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Abstract

Polyaniline (PANI) stands out as one of the most representative and promising organic electrochromic conducting polymers, attracting extensive research attention owing to its facile synthetic route, low raw material cost, reversible electrochemical redox behavior, outstanding environmental stability, and unique proton-coupled electrochromic mechanism. Electrochromic materials are functional substances that exhibit reversible, tunable variations in optical parameters including color, optical transmittance and reflectance under a low applied electric field, which are driven by ion insertion/extraction and electronic structure rearrangement. Compared with inorganic electrochromic materials such as tungsten oxide and molybdenum oxide, PANI possesses inherent merits of rich color switching, good mechanical flexibility, structural adjustability and low preparation cost, endowing it enormous application potential in smart windows, electrochromic displays, adaptive military camouflage, energy storage integrated devices and wearable optoelectronics.

This paper systematically reviews the research progress of PANI electrochromic materials, focusing on mainstream synthetic methods, fundamental electrochromic performance characteristics, key influencing factors, modification strategies, and practical application scenarios. The synthesis routes including chemical oxidative polymerization, electrochemical polymerization, template-assisted polymerization and novel auxiliary polymerization are elaborated with their respective advantages and limitations analyzed. The core electrochromic indexes of PANI involving optical contrast, response time, cyclic stability and coloration efficiency are summarized, together with the effects of dopants, electrolytes, temperature and humidity on material performance. Furthermore, common modification strategies including doping modification, composite compounding, nanostructure construction and surface modification are overviewed to solve the defects of pure PANI such as slow response, poor cycling durability and inferior solubility. Finally, the current application status, existing technical bottlenecks and future development trends of PANI electrochromic materials are discussed, aiming to provide systematic theoretical reference and technical support for the further optimization and industrialization of polyaniline-based electrochromic devices.

Keywords: Polyaniline; Electrochromic materials; Synthetic method; Electrochromic property; Modification; Application

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I. Introduction

With the rapid upgrading of intelligent buildings, flexible electronics, military stealth technology and new energy devices, electrochromic technology has become a research hotspot in the field of advanced functional materials in recent years. Electrochromism refers to the phenomenon that the optical properties of materials undergo stable and reversible modulation under the excitation of an external electric field, essentially originating from reversible redox reactions accompanied by embedding and de-embedding of small ions such as H^+ , Li^+ and Na^+ [1]. According to material classification, electrochromic materials are divided into inorganic metal oxides, organic small molecules and conjugated conducting polymers[2]. Inorganic electrochromic materials have high optical contrast and excellent long-term stability, but they suffer from high preparation cost, rigid texture, complicated film-forming process and poor flexibility, which restrict their application in flexible and portable devices[3]. Organic small molecular electrochromic materials have rich color variation but poor environmental stability and easy degradation under long-term electric field cycling[4]. Conducting polymers have become the preferred candidate for next-generation electrochromic

materials due to their unique conjugated π -electron structure, simple synthesis, adjustable molecular structure and excellent electrochemical activity^[5]. Typical varieties include polypyrrole, polythiophene, PEDOT and polyaniline. Among them, polyaniline has incomparable comprehensive advantages: raw aniline monomer is cheap and easy to obtain, polymerization reaction can be carried out under mild conditions, multiple reversible oxidation states bring rich color switching, and it maintains stable electrochemical performance in atmospheric environment without strict oxygen and moisture isolation^[6]. Since the redox state of PANI can be regulated by proton doping and electric field induction, it presents three typical intrinsic states: fully reduced leucoemeraldine, half-oxidized emeraldine and fully oxidized pernigraniline, corresponding to reversible color changes of pale yellow, green and purple-black respectively, which lays the material foundation for electrochromic regulation^[7].

Nevertheless, pristine polyaniline still has obvious deficiencies in practical electrochromic application: slow electrochromic response time, limited optical contrast, poor long-cycle stability, easy aggregation of molecular chains and poor solubility in conventional solvents^[8]. These shortcomings lead to the failure of pure PANI to meet the requirements of high-performance electrochromic devices. In response to the above problems, domestic and foreign researchers have optimized the microstructure and electrochemical properties of PANI by optimizing synthetic parameters, constructing nanostructures, heteroatom doping, and compounding with carbon materials or metal oxides^[9]. At present, PANI-based electrochromic materials have achieved remarkable progress in laboratory research, and gradually moved towards the prototype development of flexible smart windows, low-power electrochromic displays and infrared stealth camouflage devices.

This review systematically sorts out the mainstream synthesis methods of PANI electrochromic materials, analyzes the influence of different preparation processes on microstructure and electrochromic properties, summarizes the core performance indexes and optimization modification strategies, combs the latest application progress in various fields, and points out the existing challenges and future development directions, so as to provide a comprehensive reference for subsequent basic research and engineering application of polyaniline electrochromic materials.

II. Synthetic Methods of Polyaniline

The microstructure, morphology, molecular weight and film uniformity of PANI are directly determined by synthesis methods, which further dominate its electrochemical and electrochromic performance. At present, the mainstream synthesis technologies of PANI electrochromic materials are mainly divided into chemical oxidative polymerization, electrochemical polymerization, template-assisted polymerization, and novel auxiliary polymerization methods such as microwave, ultrasonic and solvothermal polymerization. Different methods have distinct process characteristics, controllability and applicable scenarios.

2.1 Chemical Oxidative Polymerization

Chemical oxidative polymerization is the most widely used and mature method for industrial and laboratory preparation of PANI, featuring simple operation, low equipment requirement, low cost and large-scale batch production^[10]. The basic principle is to oxidize aniline monomer to initiate polymerization in acidic medium by adding chemical oxidant, and form polyaniline powder or film through chain growth and molecular chain coupling^[11]. Key influencing factors include oxidant type, acid dopant concentration, reaction temperature, monomer concentration and system pH value.

Common oxidants include ammonium persulfate (APS), ferric chloride, potassium dichromate and hydrogen peroxide. APS is the most widely used oxidant because of its moderate oxidation capacity, no impurity metal ion introduction, mild reaction condition and high molecular weight of synthesized PANI^[12]. Reaction temperature is a critical control parameter: low temperature (0–5 °C) is conducive to slow polymerization, obtaining PANI with regular molecular chain arrangement, high crystallinity and uniform morphology; room temperature or high temperature will lead to violent polymerization, molecular chain entanglement and particle agglomeration, which seriously reduce electrochromic stability^[13]. The reaction medium must be acidic, and pH controlled at 0.5–2.0 is favorable for the formation of conductive emeraldine salt phase; neutral or alkaline environment will terminate polymerization and form non-conductive insulating PANI base^[14].

According to reaction medium types, chemical oxidative polymerization can be divided into solution polymerization, emulsion polymerization and suspension polymerization. Solution polymerization is simple and conventional, suitable for preparing bulk PANI materials; emulsion polymerization uses surfactant to form micelle micro-reactors, which can synthesize nanofibrous PANI with large specific surface area^[15]. The disadvantages of this method are uncontrollable film morphology, wide molecular weight distribution,

complicated post-treatment washing and drying process, and easy generation of waste liquid to cause environmental pressure^[16]. It is mostly used for the mass preparation of PANI powder and composite precursor materials.

2.2 Electrochemical Polymerization

Electrochemical polymerization refers to the in-situ growth of PANI thin film on the surface of conductive substrate (ITO, FTO, platinum, carbon electrode) by anodic oxidation of aniline monomer under applied electric field^[17]. This method has prominent advantages in preparing electrochromic direct-use films: one-step film formation without additional binder, firm adhesion between film and substrate, controllable film thickness and morphology by adjusting voltage, current and polymerization time, and no need for subsequent complex doping treatment^[18].

Common preparation modes include cyclic voltammetry, constant potential deposition and constant current deposition. Cyclic voltammetry polymerization is the most commonly adopted mode, which can form uniform and dense PANI film with good electrochemical reversibility by potential scanning in a fixed window^[19]. Substrate electrode material determines the application scenario: ITO/FTO transparent conductive glass is the preferred substrate for transparent electrochromic devices; carbon cloth and carbon paper are suitable for flexible electrochromic energy storage composite devices^[20]. The electrolyte is composed of acidic supporting electrolyte (HCl, H₂SO₄, p-toluenesulfonic acid) and solvent. Organic acid doping can effectively improve the solubility and environmental stability of PANI film^[21].

The limitations of electrochemical polymerization are low production efficiency, only suitable for single-sided electrode film formation, high cost of transparent conductive substrate, and difficult to realize large-area industrial batch preparation^[22]. Therefore, it is mainly applied in laboratory research and small-size high-performance electrochromic device prototype preparation.

2.3 Template-Assisted Polymerization

Template-assisted polymerization is an effective method to synthesize PANI with controlled nanostructures such as nanofibers, nanowires, nanotubes and porous networks^[23]. By using template confinement effect, the growth direction and microscopic morphology of PANI molecular chains are regulated to obtain materials with large specific surface area, short ion diffusion path and fast charge transfer rate, which significantly optimize electrochromic response speed and cyclic stability^[24]. Templates are divided into hard templates and soft templates. Hard templates mainly include anodic aluminum oxide (PAA), mesoporous silica and carbon nanotube arrays. Regular pore channel structure of PAA template can confine aniline polymerization in pores, and PANI nanotubes or nanowires can be obtained after template etching and removal^[25]. Soft templates adopt surfactants (CTAB, SDS), macromolecular polymers and biomolecular micelles as micro-reactors, which avoid complicated template removal process and have low cost and mild reaction conditions^[26]. Template method can precisely construct ordered nanostructured PANI, but the preparation and removal process of hard templates are complicated and time-consuming, limiting its large-scale application^[27]. It is mainly used for the preparation of high-performance nanostructured electrochromic materials.

2.4 Novel Auxiliary Polymerization Methods

In recent years, with the development of material preparation technology, microwave-assisted, ultrasonic-assisted and solvothermal polymerization have been gradually applied to the synthesis of PANI^[28]. Microwave polymerization relies on microwave uniform heating effect to shorten polymerization time from several hours to tens of minutes, with high reaction efficiency and uniform product particle size^[29]. Ultrasonic polymerization uses ultrasonic cavitation to disperse monomers and oxidants, inhibit particle agglomeration, and is conducive to the preparation of dispersed PANI nanofibers^[30]. Solvothermal polymerization conducts reaction in high-pressure sealed kettle, which can improve the crystallinity of PANI and regulate spherical and flower-like special morphology^[31]. These novel methods have obvious advantages in optimizing morphology and shortening reaction time, but they rely on special equipment and have high production cost, which are still in the laboratory research stage.

III. Electrochromic Properties and Influencing Factors of Polyaniline

Electrochromic performance evaluation of PANI mainly includes core indexes: color switching range, optical contrast, response time, cyclic stability and coloration efficiency. These properties are determined by the intrinsic molecular structure and redox mechanism of PANI, and also regulated by dopant type, electrolyte

environment, temperature and humidity.

3.1 Intrinsic Electrochromic Characteristics

PANI has three reversible oxidation states under electric field drive: leucoemeraldine (fully reduced, pale yellow, insulating), emeraldine salt (half-oxidized, green, conductive after proton doping), pernigraniline (fully oxidized, purple-black, insulating)^[32]. The reversible transition among the three states realizes multi-color electrochromic switching, which is far richer than single-color inorganic electrochromic materials. Optical contrast refers to the transmittance difference between colored state and bleached state, which is the key index to evaluate color switching visibility. The optical contrast of pure PANI film in visible light region is mostly 30%–70%^[33]. Response time includes coloring time and bleaching time, determined by ion diffusion rate and interface charge transfer resistance. The response time of pristine PANI is generally 1–10 s, which is slower than inorganic oxide materials^[34]. Cyclic stability reflects the service life of the device. Pure PANI is prone to molecular chain fracture, doping ion loss and film shedding after repeated redox cycles, and its performance decays significantly after hundreds of cycles^[35]. Coloration efficiency of PANI is 100–300 cm²/C, higher than most inorganic electrochromic materials, showing excellent energy-saving potential^[36].

3.2 Key Influencing Factors

Dopant is the core factor affecting PANI performance. Inorganic acid dopants (HCl, H₂SO₄) have high doping efficiency and improve conductivity obviously, but doping ions are easy to lose during cycling^[37]. Organic acid dopants such as camphorsulfonic acid and p-toluenesulfonic acid have large molecular volume, stable doping effect, and can improve the solubility and flexibility of PANI^[38]. Metal ion doping can adjust infrared emissivity of PANI and expand its application in military camouflage^[39]. Electrolyte determines ion migration rate and electrochemical reaction window. Aqueous electrolyte has low cost and fast H⁺ diffusion, suitable for conventional electrochromic devices; ionic liquid electrolyte has non-volatility and wide potential window, which can greatly improve the cyclic life of PANI devices up to more than 10,000 cycles^[40]. Ambient temperature and humidity also affect performance: appropriate temperature rise accelerates ion diffusion and shortens response time, but excessive temperature will destroy conjugated molecular chains; too high or too low humidity will reduce conductivity and cycling stability of PANI^[41].

IV. Modification Strategies of Polyaniline Electrochromic Materials

To overcome the defects of pure PANI such as slow response, poor cycle stability and low optical contrast, researchers have developed multiple modification strategies, mainly including doping modification, composite modification, nanostructure construction and surface modification.

4.1 Doping Modification

Doping modification introduces foreign ions or functional groups into PANI molecular chains to adjust electronic structure and microscopic morphology. Besides conventional inorganic and organic acid doping, carbon material doping (graphene, carbon nanotubes) can build conductive networks, accelerate charge transfer, and improve optical contrast and response speed^[42]. Conducting polymer doping compound PANI with PEDOT and polypyrrole to expand color switching range and enhance mechanical flexibility^[43].

4.2 Composite Modification

Compounding PANI with inorganic metal oxides (WO₃, TiO₂, V₂O₅) can combine the high stability of inorganic materials with the rich color of PANI, effectively improve cyclic life and optical contrast^[44]. Compounding with flexible macromolecules such as PVA and polyimide can significantly enhance the bending resistance and substrate adhesion of PANI film, which is suitable for flexible electrochromic devices^[45]. PANI/MXene composite has ultra-fast response and high specific capacitance, realizing integration of electrochromism and energy storage^[46].

4.3 Nanostructure Construction

Constructing nanofiber, nanowire and porous network nanostructures of PANI can increase specific surface area, shorten ion diffusion path, reduce interface resistance, and realize rapid electrochromic response within

0.5 s^[47]. Ordered nanostructures can also alleviate molecular chain agglomeration and improve long-cycle structural stability^[48].

4.4 Surface Modification

Plasma treatment and silane coupling agent modification are adopted to introduce active groups on PANI surface, enhance the bonding force between film and substrate, avoid film shedding during cycling, and further improve device durability^[49]. Surfactant surface modification can improve the dispersion and solubility of PANI, which is convenient for solution processing and large-area film formation^[50].

V.Applications of Polyaniline Electrochromic Materials

5.1 Smart Windows

PANI electrochromic smart windows can adjust visible light and solar radiation transmittance by voltage regulation, reduce building air conditioning energy consumption, and realize intelligent light and heat management. With low cost and flexible film-forming ability, it is suitable for intelligent buildings, vehicle skylights and aircraft window glass. Flexible PANI composite films can be applied to curved surface smart window scenarios, showing broad commercial prospect.

5.2 Electrochromic Displays

PANI electrochromic displays have the advantages of low power consumption, wide viewing angle, no backlight and good flexibility, which are applicable to electronic paper, wearable device display screens and smart label cards. By patterning PANI film matrix, low-cost monochrome and multi-color display can be realized, which has become a competitive candidate for next-generation flexible low-power display technology.

5.3 Military Adaptive Camouflage

PANI can realize synchronous regulation of visible color and infrared emissivity under electric field drive. It can adapt to the background color of grassland, desert and forest in visible light, and adjust infrared radiation characteristics to avoid infrared detector recognition. It can be prepared into flexible camouflage fabrics and military protective materials, and has important application value in battlefield stealth and thermal infrared camouflage.

5.4 Energy Storage Integrated Devices

PANI has both electrochromic characteristics and pseudocapacitive energy storage performance. The prepared electrochromic supercapacitor and electrochromic battery can realize real-time visual display of charge and discharge state through color change, integrating energy storage, display and regulation functions. It is widely used in wearable electronic devices and self-powered intelligent optoelectronic systems.

5.5 Other Applications

PANI electrochromic materials can also be applied in optical switches, electrochromic sensors, anti-glare rearview mirrors and aerospace thermal control devices. With the continuous optimization of modification technology, its application boundary is further expanding.

VI.Result and conclusion

This review comprehensively summarizes the research progress of polyaniline (PANI) electrochromic materials, focusing on their intrinsic laws and application bottlenecks from synthesis, performance, modification and application aspects, and discusses core results and existing problems.

In terms of synthesis, there is no universal optimal route, and the selection depends on application scenarios and performance requirements. Chemical oxidative polymerization is mature and suitable for large-scale production but has defects in morphology control; electrochemical polymerization is ideal for laboratory-prepared high-performance films but is inefficient and costly; template-assisted polymerization can fabricate nanostructured PANI but is limited by complex template processes; novel auxiliary methods shorten reaction time but rely on special equipment. Future synthesis research should focus on integrating multiple methods to achieve low-cost, large-scale preparation of high-performance PANI.

In terms of electrochromic performance, PANI has unique multi-color switching and high coloration efficiency (100–300 cm²/C), but pure PANI suffers from low optical contrast (30%–70%), slow response time (1–10 s) and poor cyclic stability. Dopants, electrolytes, temperature and humidity can regulate performance but cannot fundamentally solve inherent defects.

Modification is key to overcoming PANI's defects. Synergistic modification is more effective than single strategies: carbon/conducting polymer doping improves conductivity and response speed; compounding with inorganic oxides enhances stability and contrast, while compounding with flexible macromolecules boosts flexibility; nanostructure construction shortens response time; surface modification improves substrate adhesion. PANI/MXene composites are a research hotspot, but modification technology still needs optimization for complexity, cost and reproducibility.

In applications, PANI shows broad prospects in smart windows, displays, military camouflage and energy storage devices, but each field faces specific bottlenecks (e.g., slow response for smart windows, low resolution for displays) that need to be addressed for commercialization.

Current industrialization bottlenecks include insufficient cyclic stability of large-area films, inadequate performance, immature film-forming/packaging technologies and poor compatibility. These interrelated issues require synergistic optimization of synthesis, modification and device preparation. In summary, PANI has significant advantages and research progress, but technical bottlenecks remain; integrating synthesis methods, developing efficient modification technologies and accelerating lab-to-industry transformation are key to its future development.

VII.Challenges and Future Perspectives

Although PANI electrochromic materials have made great progress in synthesis, performance optimization and application research, there are still many bottlenecks restricting industrialization: first, the long-term cyclic stability of large-area PANI film is insufficient, and it is easy to age and fail in atmospheric environment; second, the response speed and optical contrast still need to be further improved to meet high-end display and camouflage requirements; third, the large-area uniform film-forming technology and flexible device packaging technology are immature; fourth, the compatibility between PANI and flexible substrate and solid electrolyte needs to be optimized.

Future research directions should focus on: (1) optimizing green and low-cost synthesis and large-area uniform film-forming technology; (2) developing multi-component composite and nanostructure synergistic modification system to comprehensively improve electrochromic response, contrast and cycle life; (3) designing high-stability solid electrolyte and flexible packaging materials to adapt to flexible wearable devices; (4) promoting the engineering prototype development of PANI smart windows and military camouflage devices, and accelerating the transformation from laboratory research to commercial application; (5) exploring the multifunctional integration of electrochromism, energy storage, sensing and thermal control to expand the application value of PANI materials.

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