Review Article

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Application of melanin as biological functional material in composite film field

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Abstract: Melanin comes from a wide range of sources. It can be isolated and characterized from some plants, animals and microorganisms, and can also be simply polymerized by dopamine. It has many biological properties such as antioxidant, ultraviolet shielding and bacteriostasis. Based on the above characteristics, the addition of melanin to film-forming materials can improve the relevant physical properties and functional properties of the film. In this article, the structure and properties of melanin were briefly introduced, and then the advantages and related research progress of melanin as a functional filler in the field of composite film were summarized.

Keywords: melanin, functional filler, composite film, excellent performance, food packaging, photonic devices

1 Introduction

Compared to petroleum-based synthetic plastics, biopolymer-based films are biodegradable and have good biocompatibility [1–4]. At present, researchers have confirmed by X-ray diffraction (XRD), scanning electron microscope (SEM) and other characterization methods that adding functional fillers can improve the physical properties and functional properties of the films [5–8]. Melanin is a kind of biological macromolecule widely existing in the human body, with ultraviolet (UV) radiation shielding, antioxidant and antibacterial properties. It also has strong adhesion and can adhere to almost all substances [9]. Its active group can react with target molecules, making it easier to achieve surface modification of substances [10]. Due to these characteristics of melanin, researchers began to combine melanin with different materials such as polycarbonate (PC) and polyvinyl-alcohol (PVA) to prepare composite films, which have more excellent properties [11–15]. At present, melanin composite film has been widely used in food packaging, biomedicine, electronic devices and other fields [16–19]. As shown in Figure 1, the properties of melanin and its application as functional materials in composite films are reviewed in this article. This will not only provide a theoretical basis for the preparation of composite membranes with better performance, but also help to optimize the allocation of melanin resources.

2 Overview of melanin

2.1 Structure and classification of melanin

At present, there is no uniform standard for the classification of melanin in the world. Nicolaus divided melanin into eumelanin, brown melanin and isomelanin. According to different sources, melanin can be divided into animal melanin, plant melanin and microbial melanin [20,21]. According to different preparation methods, melanin can be divided into synthetic melanin and natural melanin [10].

There are many kinds of melanin in nature, and the most widely studied are eumelanin and brown melanin, which have a common dopa quinone precursor. Eumelanin is brown or black and is formed by oxidative polymerization of 5,6-dihydroxyindole (DHI) and 5,6-dihydroxyindole-2-carboxylic acid (DHI-CA) monomers. Brown melanin is yellow; it is oxidized to dopa quinone by tyrosine, and then cyclized and decarboxylated by cysteine [22–25]. Natural melanin is insoluble in water and conventional organic solvents, so it is difficult to conduct physical and chemical analysis of it, so its precise chemical structure has not been determined [10].
In recent years, scientists have begun to oxidize and polymerize dopamine in alkaline solution to form melanoid polydopamine nanoparticles, which have the same physical and chemical properties as natural eumelanin [26,27]. So the melanoid produced by dopamine is also called dopamine melanin or polydopamine [28–30]. After continuous development, related studies on melanoid polydopamine have been increasing year by year [31]. Scholars proposed the melanoid eumelanin model based on the formation pathway of natural eumelanin in vivo [32]. They believed that dopamine first forms quinones, and then DHIs are obtained through cyclization and rearrangement, and DHIs are easily oxidized to 5,6-indole quinones. Both DHIs and 5,6-indole quinones branched at sites 2, 4 and 7 to form oligomer isomers with different degrees of polymerization. Finally, melanoid was obtained by counter disproportionation reaction.

2.2 Characteristics of melanin

2.2.1 Antioxidant activity

The phenolic structure in the melanin molecule is the main electron-donor group, which is responsible for antioxidant activity [33,34]. Liu et al. detected by electrochemical method that melanin has redox properties and will repeatedly switch between oxidation state and reduced state [35]. In addition, melanin can also quickly capture the alkyl, alkoxy and peroxide free radicals produced when the polymer chain is broken. The reducing ability of melanin and the ability to remove hydroxyl free radical superoxide anion in the seed coat of Apricot are better than that of Vitamin C (Vc) [36]. Melanin in the body of Daihe Silky chicken has a better ability to remove 1,1-diphenyl-2-picrylhydrazyl free radical (DPPH) [37]. Black auricularia melanin has obvious scavenging effect on superoxide anion hydrogen peroxide and other free radicals, and also has a good protective effect on deoxyribonucleic acid (DNA) damage [38]. At the same concentration, the scavenging effect of Aureobasidium pullulans melanin on hydroxyl radical, superoxide anion free radical and DPPH free radical is higher than that of Vc [39].

2.2.2 Ultraviolet (UV) shielding

Melanin has broadband absorption capacity to visible and UV light. Based on the characteristics of melanin, it realizes ultraviolet shielding by absorbing and partially scattering ultraviolet light. Huang et al. made melanoid nanoparticles form a miniature “solar umbrella” in the epidermal keratinocytes of organisms to protect DNA from ultraviolet radiation [40].
2.2.3 Other properties

Melanin is currently the only known endogenous anti-radiation substance that can avoid ultraviolet, X-ray, γ-ray and other damage to cells in the body [41,42]. *Trichococcus granulosus* melanin can significantly improve the survival rate of *Staphylococcus aureus*, *E. coli* and *Saccharomyces cerevisiae* under ultraviolet radiation. Anti-radiation experiments in mice also showed that melanin has strong anti-ultraviolet radiation activity [43]. Moreover, melanin contains a variety of functional groups, such as C==O, COOH, –OH, etc., which can provide multiple non-equivalent binding sites for metal ions [44]. After chelating with metals such as calcium, iron and zinc, many functions of animals and plants can be improved [45,46]. Melanin is also an excellent semiconductor material with strong light absorption capacity in the near infrared region, which has been widely used in electronics and bioactive compounds and other fields [47,48]. Table 1 lists some characteristics and related mechanism of melanin.

2.3 Melanin composite film

The problems of non-degradable plastic packaging materials are increasing. The packaging of biomolecular materials is environmentally friendly and has excellent performance. The films made of carrageenan gelatin and other materials had been widely used in packaging and other fields, but they also have their own disadvantages. Previous studies have confirmed that the addition of functional fillers can improve the physical and functional properties of the film [5–8] and the biocompatibility and degradability of organic fillers are superior to inorganic materials. Melanin, as a crosslinking agent, can improve the physical properties of biopolymer and give it functional properties such as antioxidant, antibacterial and UV shielding [50]. In this context, it is very attractive that melanin as a functional filler enhances the performance of composite film. At present, melanin has been gradually used as a functional filler to add PVA and poly(lactic acid) (PLA) to prepare melanin composite films [15,51,52] (Table 2).

3 Melanin as a functional filler to improve the performance of composite film

The properties of the film include physical and chemical properties and separation and permeability properties. The former mainly refers to oxidation resistance, mechanical strength and hydrophobicity. The latter mainly refers to the osmotic flux and separation efficiency. A large number of catechol structures in melanin can interact with the matrix, which greatly improves the performance of the composite materials. Therefore, researchers have successively used melanin as a functional filler to improve the mechanical strength and antioxidant properties of the film.

3.1 Antioxidant activity

Melanin molecule with oxidizing (adjacent quinone) and reducing (adjacent hydroquinone) group, can interact with free radicals, losing in the process of electron transfer electronic or capture to remove active oxygen or reactive nitrogen free radicals. Another possible synergy between the different functional groups make melanin oxidation resistance stronger [35,36]. The results showed that

| Characters                 | Mechanism                                                                 | References |
|----------------------------|---------------------------------------------------------------------------|------------|
| Antioxidative              | The phenolic structure of the melanin molecule is the main electron-donor group responsible for antioxidant activity so it can seek out and remove excessive free radicals in the organism | [33–39]    |
| Ultraviolet shielding      | It has broadband absorption capacity for visible and ultraviolet light    | [40]       |
| Anti-radiation             | It is the only known endogenous anti-radiation substance that can avoid damage to cells in the body caused by ultraviolet, X-ray, γ-ray and ionizing radiation | [41–43]    |
| Chelating metal            | It contains a variety of functional groups, such as –C==O, –COOH, –OH, and can provide multiple non-equivalent binding sites for metal ions | [44–46]    |
| Photothermal conversion    | It has strong light absorption capacity in the near infrared region       | [47,48]    |
| Anti-bacteria              | It can significantly reduce biofilm formation of food pathogens           | [49]       |

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Table 2: Advantages and disadvantages of commonly used film forming materials

| Materials         | Advantage                                                                 | Disadvantage                                                                 |
|-------------------|---------------------------------------------------------------------------|-------------------------------------------------------------------------------|
| Carrageenan       | Strong gel-forming ability, biocompatibility, high environmental friendliness and reproducibility | Mechanical and water vapor barrier performance is weak                          |
| Gelatin           | Wide range of sources                                                     | Poor mechanical, heat and water vapor barrier performance                      |
| Chitosan          | Highly biocompatible and degradable                                        | Flexible mechanical properties and weak barrier performance                     |
| Agar              | Good film formation and good gel formability                              | Poor mechanical properties and water barrier performance                       |
| Cellulose         | Wide source, renewable, easy chemical modification, non-toxic, excellent mechanical and optical properties | Weak water vapor resistance and high hydrophilicity                            |
| Polylactic acid   | Degradable, biocompatible high bacterial resistance, flame retardancy and UV resistance | Thermal stability, impact resistance, solvent resistance and other properties are poor |
| Polyethylene      | Low temperature resistance, chemical stability and high electrical insulation | Easy to degrade under ultraviolet light                                          |
| Polystyrene (PS)  | Radiation resistance and electrical insulation                             | Easy to light oxidation, thermal oxidation, ozone decomposition and UV degradation |
| Poval             | Good compactness, strong adhesion, solvent resistance, wear resistance and high gas resistance | Complex process, high cost and low output                                        |
| Polycarbonate     | Good transparency, excellent mechanical properties, oil and acid resistance | UV resistance, poor wear resistance, not resistant to strong alkali and oxidizing acid and weak UV resistance |
| Polyurethane      | Good elasticity, excellent water proofing, cold resistance, environmental protection and non-toxic | Weak durability, many micro-voids, low tensile strength and easy to crack under external force |

melanin can be used as a functional filler to develop anti-oxidant biopolymer film.

Roy et al. added melanin nanoparticles (MNP)s to Agar and chitosan, respectively, to prepare nano-composite film with strong oxidation resistance [12,53]. Lopusiewicz et al. prepared a composite film with good antioxidant activity by adding fungus-derived melanin to PLA [51]. Dong et al. found that melanin could capture free radicals produced during PVA degradation [54]. Phua et al. found that the nanofilm coated with melanin layer can act as a free radical scavenger to inhibit thermal and oxygen aging of polypropylene [55].

3.2 Photothermal stability

The light stability of a material refers to its resistance to light aging, while thermal stability refers to the heat resistance of the material. Dong et al. found that the addition of melanin to film forming materials can improve the photothermal stability of the film. The UV shielding performance and transparency of polycarbonate melanin composite film are related to the size of melanin, and reducing its particle size can improve the UV shielding efficiency and transparency. This is because the smaller the particle size of melanin is, the larger the specific surface area is, and the larger the effective area for ultraviolet absorption and heat conversion is [54].

Roy and Rhim prepared carrageenan-based nanocomposite film by solution casting method. The results showed that the addition of MNPs completely screened UV light and improved the heat resistance of the film [56]. Roy and Rhim prepared AgNPs with melanin as reducing agent and combined with carrageenan to prepare composite film [16]. Wang et al. combined melanoid and PC to improve the photothermal stability and anti-aging properties of the film [14]. Liang et al. found that the alpaca melanin coating enhanced the light stability of wool fabric [57,58]. Xie et al. prepared a composite film with good photostability by embedding melanin nanoparticles/TiO₂ into polyurethane [52]. Kablov et al. prepared a composite film containing grape seed extract (GSE) by extrusion and blow molding method. The results of low density polyethylene (LDPE)-based composite film of melanin nanoparticles and zinc oxide nanoparticles (ZnONPs) showed that the thermal stability of the composite film is improved [59].

3.3 UV shielding performance

UV shielding refers to the ability to reflect or absorb ultraviolet light. Studies have shown that melanin can be used...
as a functional filler to improve the UV shielding performance of the film forming materials. It is expected to be used as UV shielding film in food packaging and cosmetics industry. In recent years, many substances have been reported to improve the UV shielding performance of polymers [60,61]. Organic UV absorbers such as benzophenone can disperse UV, but their embedding in polymers often causes photodegradation migration or aggregation. Inorganic oxides, such as silica, weaken UV rays through band gap absorption and scattering of light [62,63], but they can cause photodegradation of polymer matrix [15,64]. Graphene and melanin are currently the most commonly used organic UV absorber [65–68]. Melanin has a wide range of ultraviolet absorption ability, can block ultraviolet light and reduce UV transmission effect, which is the most promising candidates. Melanin is UV resistant because it quickly converts photon energy from UV light into heat to prevent photodegradation damage [13,69]. Wang and Xue found that melanin has obvious short-wavelength shielding and high visible light transparency, and its load rate of 1% will block most UV light [13]. Therefore, researchers began to add melanin to the film forming materials to improve the UV resistance of the film.

Wang et al. prepared polydopamine hollow nanoparticles (PDA-H NPs), and then prepared PDA/PVA nanocomposite film. The results showed that the prepared nanoparticles could quickly convert the energy of UV light into heat energy, and the composite film had good UV shielding performance [15]. Di Mauro et al. found that eumelanin increased the UV absorption of polyethylene-based film by one order of magnitude [70]. Roy et al. prepared chitosan/MNPs nanocomposite film and the results showed that MNPs enhanced the UV barrier of the composite film [71].

3.4 Antibacterial activity

Bacteriostasis is the use of physical or chemical methods to kill or inhibit the growth and reproduction of microorganisms. Laxmi et al. found that melanin achieved its antibacterial activity against Vibrio parahaemolyticus and Staphylococcus aureus mainly by inhibiting or damaging the formation of biofilm, increasing the leakage of cell contents and decreasing the cell film potential [72]. Xu also found that melanin significantly inhibited bacterial biofilm formation [73]. Therefore, some studies have added melanin to the film forming materials to improve the antibacterial performance of the film, and broaden the application of the film forming materials in the packaging materials to avoid bacterial contamination.

Roy et al. prepared AgNPs with melanin as reducing agent, and combined with carrageenan to prepare composite film with strong antibacterial activity against E. coli [50]. The melanin/TiO\(_2\) composite synthesized by Vitiello et al. showed significant antibacterial activity [74], Kiran reported that the nano-composite film prepared by the blend of polyhydroxybutyrate, nano-melanin (PHB:Nm) and glycerol had strong antibacterial activity [75]. Both the melanin carrageenan-based composite film prepared by Roy and Rhim [16] and the LDPE nanocomposite film prepared by Kablov et al., which contained GSE, melanin and ZnONPs, showed strong antibacterial activity [59]. Su et al. reported for the first time the preparation of strong antibacterial PDA paint by shaking method. The results showed that the antibacterial activity of the PDA coating was significantly enhanced [76].

3.5 Electrical conductivity

Conductivity refers to the ability of an object to conduct current. A conductive film is a film that conducts electricity. Over the years, many materials have been proposed for the development of electronic devices, but all have certain disadvantages [77,78]. Poly 3,4-ethylenedioxythiophene-polystyrene sulfonate (PEDOT:PSS) is hygroscopic [79] and graphene cannot be used on large area substrates [80]. The conductivity of polyaniline (PANI) is not high [81] and the dispersion of metal nanoparticles or carbon nanotubes (CNTs) in conductive polymers reduces the transparency [82]. A key problem that still needs to be solved is the development of a good conductive organic material [83]. Studies have shown that the melanin coating on the surface of other materials can partially improve the conductivity [84]. With the ability to carry electron and ionic charges, these findings provide an opportunity for emerging applications of implantable green bioelectronics.

Some studies have found that mixing PEDOT:PSS (commonly used conductive material) with DHI improves the cohesion, water resistance and electrical conductivity of the film, which can be used as the anode material of organic LED [85]. Eom et al. prepared a composite material with an electrical conductivity of 1.17 ± 0.13 S cm\(^{-1}\) at room temperature by using MNPs derived from squid ink and PVA, which is the best composite material for preparation of biological nanoparticles at present [86]. After oxidizing hydroxyl groups on the surface of cellulose nanocrystals to carboxyl groups, Chen and Huang added PDA coated and reduced graphene oxide nanoparticles (PDA-rGO), and prepared a composite film with significantly
improved electrical conductivity by casting film formation method [87].

3.6 Other performance

The addition of melanin as a functional filler can not only improve the above properties, but also significantly improve the hydrophobic mechanical properties and biodegradability of the film. Yi and Shen prepared superhydrophobic composite film by drop-casting PS@PDA NPs and dipping a layer of polydimethylsiloxane (PDMS) on the film [88]. Melanin is one of the important biological materials for green synthesis of CuO NPs [89–92]. Roy et al. prepared copper oxide nanoparticles (CuO) with melanin as stabilizer. The Agar/CuO NPs composite film were prepared by solid solution method. The results showed that the surface hydrophobic properties of the composite film are improved [93]. Dong et al. prepared the composite with natural melanin and PU with significantly improved mechanical properties. The tensile strength and elongation at break are increased from 5.6 MPa and 770% to 51.5 MPa and 1,880%, respectively, and the toughness is increased by 10 times [94]. Dong et al. significantly improved the mechanical properties of the polymer through the hydrogen bonding between synthetic melanin nanoparticles and PVA molecules [95]. Shankar et al. found that melanin can significantly improve the mechanical strength of composite films [96]. Roy et al. prepared the cellulose nanofiber CNF/MNPs composite film and the results showed that MNPs enhanced the mechanical properties of the film [97]. Xing et al. prepared composite film by combining PDA with lignin nanoparticles (LNP) and polybutylene terephthalate (PBAT). The results showed that the functional groups such as –OH and –COOH of melanin formed strong hydrogen bonds with the polymer chain containing polar groups, which significantly improved the mechanical properties of the composite material [98]. Eom et al. prepared natural MNPs/PVA composite film and the results showed that MNPs added to PVA could significantly improve the appetite of super worms [99].

Compared to the current literature reports, the addition of melanin as a functional filler to the film forming material has a relatively high enhancement effect on the film performance. The use of melanin can obtain a degradable composite film with excellent superhydrophobic and mechanical properties, which has a good potential in packaging or agricultural applications.

4 Progress in the application of melanin in composite film

Based on the above results, melanin can be used as a functional filler to improve the physical and functional properties of the film, and broaden the application of film forming materials. Melanin composite film have been widely used in food packaging materials to avoid the effects of light irradiation and temperature. In fact, it also has a broad application prospect in biomedical and optical materials.

4.1 Food packaging

Microbial light (especially UV light) and oxygen in the air are the main factors that promote food aging and deterioration. Therefore, antioxidant ultraviolet shielding and antibacterial properties are important properties of food packaging film.

Roy et al. found that compared with pure carrageenan, melanin composite film has improved water vapor barrier and thermal stability, and has stronger antibacterial activity against \textit{E. coli} [16]. Bang et al. prepared polypropylene (PP) and melanin composite film by extrusion casting method, which can prevent ultraviolet rays from penetrating and effectively prevent potatoes from turning green due to chlorophyll production [17]. Roy and Rhim prepared nanocomposite film with high antioxidant activity using MNPs and Agar [53]. Kiran et al. prepared a nano-composite film with strong antibacterial activity and heat resistance by blending PHB with melanin [75]. Lopusiewicz et al. isolated melanin from watermelon seeds and used it as a modifier of whey protein concentrate and separation film (WPC and WPI). The results showed that the melanin modification enhanced the UV blocking, water vapor blocking and antioxidant activities of WPC/WPI film [100].

The above results indicated that more and more studies have been conducted on the preparation of biodegradable film by combining melanin and film forming materials, which can be used to enhance the antioxidant, antibacterial and UV shielding properties of composites. High UV protective packaging film with melanin can be used in active packaging applications to prevent photodegradation and maintain the quality and prolong the shelf life of packaged foods. Functional biopolymer film prepared by adding melanin as a functional filler to film forming materials have broad application prospects in food packaging.
4.2 Biological medicine

Manini et al. reported for the first time that the prepared melanin film had super smooth morphology and good biocompatibility, which opened up a new way for its application in the field of tissue engineering and regenerative medicine [101]. Currently, melanin is mainly added to the film forming materials to prepare hydrogel composite film for wound healing.

Infection is a major obstacle to wound healing. Wound dressings are important materials to recover skin wounds. Hydrogels with antioxidant and antibacterial properties have become dressings to accelerate wound healing. Tao et al. prepared a hydrogel film with PDA nanoparticles and gelatin, which has good biocompatibility and can effectively bind the pore-forming toxins (PFTs) that attack host cells [102]. Liang et al. prepared gelatin-grafted dopamine (GT-DA)/chitosan/polydopamine coated carbon nanotubes (CNT-PDA) composite hydrogel, which has good biocompatibility, photothermal effect and antibacterial activity, and has a good therapeutic effect on mouse skin [18]. Li et al. prepared a hydrogel by using PDA-rGO and poly(N-isopropylacrylamide). The hydrogel has thermal response, self-shrinking tissue adhesion, anti-infection, anti-oxidation and other functional properties [103]. Li et al. prepared unidirectional water-conducting composite film by mixing silk fibroin (SF) and polycaprolactone (PCL) on PVA film surface treated by PDA. The results showed that the composite film was non-toxic and had a good wetting gradient, which was conducive to the directional transfer of water [104]. Gao et al. prepared a hydrogel film by photo-induced polymerization using PDA and rGO. It attaches to the wound and converts sunlight into heat to increase the local temperature of the wound, which then reduces the inflammatory response and promotes wound healing [105]. Wang et al. applied PDA to antheraea pernyi silk fibroin (AF) film to prepare polydopamine-coated AF (PAF film) and found that PDA coating significantly improved the roughness and hydrophilicity of the composite film, thus improving the protein absorption capacity [106] (Figure 2). Fu et al. prepared a hydrogel with good antioxidant and antibacterial properties by combining reduced PDA nanoparticles (rPDA NPs) with chitosan [107].

Taken together, these studies demonstrate the great potential of melanin composite hydrogel film for clinical or daily wound healing, opening up a new avenue for the development of antibacterial dressings.

4.3 Bioadsorption

Bioadsorption is the phenomenon that substances are adsorbed on the surface of organisms through covalent electrostatic or molecular force action. Adding nanomaterials to polymer film is an effective strategy to improve the seawater desalination performance of the film. Song et al. prepared a composite film after adding PDA nanospheres to cellulose triacetate (CTA). The results showed that the rich hydroxyl group of the nanosphere helps to improve the hydrophilicity of the film and enhances the water flux. Due to the synergistic effect of size repulsion and electrostatic repulsion, the rejection rate of salt is high (up to 90% in most cases), and the rejection effect of monovalent and divalent ions is good [108] (Figure 3). Park et al. prepared PDA film that could better separate Mg$^{2+}$ and Li$^+$ in solution [109].

4.4 Photonic devices

Clulow et al. revealed that nearly half of melanin protons are unstable, so it can be used for charge conduction. This result, combined with potential biocompatibility, makes melanin an excellent material for film bioelectronic devices [110].

4.4.1 Biosensors

Sensors such as pH and humidity have wide applications in industrial medical diagnosis and other fields [111]. Silva et al. studied the pH sensing characteristics of the extended gate field effect transistor (EGFET) with indium
tin oxide (ITO) and gold as the substrate and melanin film as the active layer. The pH range is 2–12 and the sensitivity is between 31.3 mV/pH and 48.9 mV/pH [19]. Tehrani et al. prepared a graphene-based and melanin-based pH sensor through the screen printing process that has high stability and superior performance compared to many existing devices [112] (Figure 4). Wu et al. prepared a kind of dopamine melanin film, which formed and self-assembled into a humidity sensor at the air/solution interface. The results showed that the film had good ultra-sensitivity and repeatability, with good response (5–7 s) and recovery (6–9 s) time [113].

These results indicated that the melanin composite film is a promising sensing material. The combination of high biocompatibility and high electrical conductivity makes this kind of film have a promising application in bio-electronic devices such as electronic products biosensors and implantable devices.

### 4.4.2 Height colorimetric

Non-compact colloidal crystal arrays (NCCAs) have flexible structure and adjustable color, but most of them are limited by approximate refractive index or high background scattering, which hinder their practical applications. Melanin particles produce structural colors on bird feathers, providing inspiration for the development of

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**Figure 3:** (a) Schematic diagram of the synthesis of dopamine-melanin nanospheres, (b) CTA and nanocomposite film loaded with different dopamine-melanin nanospheres, (c) water flux and interception rate of NC-0.4 film for MgSO₄, Na₂SO₄, MgCl₂ and NaCl solution and (d) separation mechanism of nanocomposite film [108].

**Figure 4:** (a) Top view of electrode, (b) optical image of flexible sensor and (c) electrode section of pH sensor based on graphene and pigment melanin (PGM) [112].
multifunctional photonic devices. Melanin is a rare material with high refractive index and high absorption, and has good adhesion and film forming properties. Based on these properties, researchers tended to add melanin to the film forming materials to improve the related properties of the film.

Liu et al. dispersed melanin particles in polydihydroxyethyl methacrylate (pHEMA) to prepare highly colorimetric film with NCCAs [114]. Yi and Shen assembled polystyrene coated core–shell nanoparticles (PS@PDA NPs) with 3-aminopropyl methoxysilane (APTES) through a heat-assisted self-assembly method. Subsequently, crack-free amorphous PCs with enhanced chromatic aberration and low angle dependence were prepared [115] (Figure 5). Xiao et al. demonstrated the change in structural color of self-assembled SMNP film in response to humidity change. This process is driven by the hygroscopic property of the particles [116]. Kohri et al. used PDA to prepare background and angle-dependent structural color film [117]. Zhu et al. prepared a non-rainbow structure color film on the surface of a silk fabric with DA. The surface of the color fabric has a double layer structure. One layer is an extremely smooth film, and the other is PDA spherical particles. The color of the fabric structure depends on the thickness of the film. As time goes on, the thickness of the film increases, and the colors are successively yellow, red, blue and green. When the thickness reaches a certain value, the color of the fabric structure is periodic [118]. Bai et al. prepared film by self-assembling PDA coating particles and found that PDA coating could not only control the color brightness/angle dependence of photonic film, but also significantly improve the color quality [119].

Since PDAs are highly adherent to almost all types of surfaces, it is possible to extend this PDA-based approach to a variety of different colloidal particles, leading to the development of photonic devices that can be widely used in the design of a variety of color smart sensors and soft devices.

### 4.4.3 Thermoelectric devices

Efficient solar energy collection and thermal energy conversion is the goal of modern sustainable energy. Based on melanin’s excellent solar-thermal conversion performance and its non-biological toxicity, researchers are now trying to apply melanin to the field of thermoelectric devices.

Studies by Wang et al. showed that the combination of melanin and halide perovskite light absorbance has a high absorption rate in the UV to near-infrared region in the solar spectrum. Under single solar light, the photothermal quantum yield reaches 99.56% and the photothermal conversion efficiency reaches 81%, which is better than graphene (70%) and other materials. By coating the photothermal composite film on the hot side of the thermoelectric device, the output power under light was increased by 7,000% [120]. Lee et al. deposited synthetic melanin film...
on the surface of various inorganic semiconductors (such as \( \text{Fe}_2\text{O}_3 \) and \( \text{TiO}_2 \)) by electropolymerization. The results showed that the deposition of complex film significantly improved the solar water oxidation performance of semiconductor photoanode [121]. Zong et al. found that PDA nanofibers showed high absorption rate and high solar heat conversion efficiency (86%) in the solar spectrum [122].

These studies use the physical and chemical properties of melanin to provide a promising platform for efficient solar energy collection, which can be used in solar evaporation, desalination and solar power generation.

### 4.4.4 Other applications

In addition to the above points, melanin has been gradually applied in capacitor quantitative detection of biological memory devices and other fields.

Albano et al. prepared PVA/synthetic melanin transparent film and found that the UV shielding film can also be used as transparent capacitor [123]. Jia et al. established gold nanoparticles/PDA-rGO composite membrane sensor. The results showed that the sensor has high sensitivity to the measurement of paraquat, and can be used for the determination of paraquat content in water and soil samples [124]. Gurme et al. prepared a functional biological memory device using melanin. Jia et al. established gold nanoparticles/PDA-rGO composite film sensor. The results showed that the sensor can be used for the determination of paraquat contents in water and soil samples [125].

### 5 Discussion

Melanin is a group of abundant, inexpensive and nontoxic materials with numerous applications due to its various physicochemical properties. Melanin can be easily extracted from multiple natural sources or chemically synthesized. Melanin has certain antibacterial activity, strong oxidation resistance and strong UV shielding performance, so it is an ideal filling material for preparing functional composite film. Composite films prepared by melanin as functional fillers have great development potential in many industrial fields such as food packaging, biomedical and electronic devices. However, more research is needed for melanin’s scale-up process, including optimized extraction and purifications of natural melanin and synthetic melanin-like materials.

### 6 Conclusion

1. Melanin is one of the ideal fillers for the preparation of functional composite film because of its wide source and non-toxic and biological properties. It is apparent that the incorporation of melanin to biopolymer or other polymer-based films significantly improves physical and functional properties of the film.

2. Comparing to other biological macromolecule materials, melanin still faces some challenges and issues: the current study is still in its infancy, more research is still needed for the mass production. But there is no doubt that melanin has many excellent properties and extremely broad application prospect and development potential in the field of composite materials.

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