



Artificial melanin particles: new building blocks for biomimetic structural coloration

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Abstract

Melanin, a black component of human hair, plays an important role in bright structural colorations in nature. For example, the beautiful and highly visible colorations of peacock feathers are achieved by periodic structures formed by melanin granules that have light absorbing capabilities. In recent years, polydopamine, which is easily obtained by the self-oxidative polymerization of dopamine, has attracted attention as a mimic of natural melanin. This focus review provides an overview of our recent research on structural color materials created using polydopamine-based artificial melanin particles and research trends in this area.

Introduction

Nature produces various coloring materials, such as dyes, pigments, and structural colors (Fig. 1). Dyes and pigments are materials that exhibit the color of reflected or transmitted light as the result of wavelength-selective absorption. Structural coloration is produced by optical interactions of light and submicron-sized periodic structures and is dependent on the size and arrangement of nanostructural elements [1]. There are several modes by which structural colors may be produced, e.g., reflection, diffraction, film interference, and scattering (Fig. 1). Depending on the application, it is necessary to select suitable periodic structures. Pigment colors may tend to fade due to the decomposition of pigment under ultraviolet light. In contrast, structural colors do not fade as long as their periodic structures are maintained. The most prominent example is the color of fossilized bird feathers. While the fossils are formed as a result of the replacement of animal tissue with minerals that are usually colorless, in rare cases, colored fossils are found [2]. These colors are reported to arise from structural coloration due to the periodic structure of the fossilized feathers. Structural colors, with unique gloss, high color developability, and considerable durability, are

expected to have applications in various fields, and numerous studies have been performed on the development of structural colors. There have been many advances in the development of structural color materials by the assembly of colloidal particles [3–11]. A structure in which mono-dispersed submicron-sized colloidal particles close to the wavelength of light are regularly arranged is called a colloidal crystal structure, which produces angle-dependent structural colors [12–14].

Well-known examples of beautiful structural colors found in nature include the blue color of the *Morpho* butterfly and the green luster of the jewel beetle. Melanin, a black material that absorbs broadband UV-visible light, plays an important role in the structural coloration of these organisms. The brilliant blue color of *Morpho* butterflies originates from the periodically arranged shelf structure, and a melanin layer exists in the lower portion of the shelf structure [15]. The jewel beetle exhibits beautiful iridescent structural color from a multilayer film in which layers of melanin and cuticle are alternately stacked [16]. Usually, when light strikes a submicron-sized structure, the structure appears white to the human eye due to light scattering. For the *Morpho* butterfly and jewel beetle, the bright structural colors appear because the melanin in the periodic structures suppresses background light. Another example of colors produced by melanin-based periodic structures is found in peacock feathers [17]. In the barbules of peacock feathers, the periodic structure of melanin granules absorbs scattered light, creating a bright structural color. In vivo, melanin is produced by multienzymatic reactions from L-3,4-dihydroxyphenylalanine (DOPA), an amino acid [18]. Thus,

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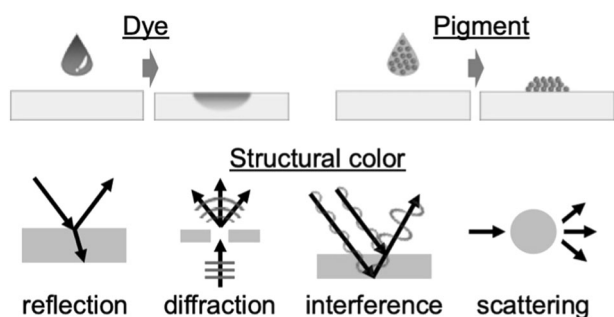


Fig. 1 Schematic illustration of three kinds of coloring materials: dye, pigment, and structural color

artificially synthesizing periodically controlled structures of melanin using biosynthetic pathways is difficult.

Recently, polydopamine, which is a polymer easily obtained by self-oxidative polymerization of dopamine under basic conditions, has attracted attention because of its ability to adhere to the surfaces of various substrates. Since the first report by Lee et al. in 2007 [19], the scope of polydopamine research has rapidly expanded [20–25]. In terms of chemical structure, polydopamine is a mimic of natural melanin (Fig. 2a, b). Research is in progress that focuses on the relationship between melanin and polydopamine from the viewpoint of structure and function [26]. Inspired by the function of melanin, several polydopamine-based functional materials have been developed, such as UV absorbers [27], antioxidants [28], and free radical scavengers [29]. As mentioned above, melanin is an important component of structural colors in nature. This focus review summarizes our recent endeavors in the creation of structural color materials using polydopamine-based artificial melanin.

High visibility structural colors from artificial melanin particles

Our research group focuses on the preparation of functional polymeric materials through the development of new methods for the synthesis and surface modification of colloidal particles [30–37]. In the course of investigating mechanisms that control the morphology of polydopamine at the particle surface [38], we found that the polymerization of dopamine in solvents composed of water/methanol mixtures produced relatively uniformly sized polydopamine particles. In 2015, we serendipitously discovered that the concentrate of an aqueous dispersion of monodisperse polydopamine particles that mimic melanin granules exhibited beautiful structural colors (Fig. 2c) [39]. These colors were produced by the scattering-based structural coloration from the assembly of submicron-sized polydopamine particles. Deep-blue, blue, green, yellow-green, orange, and red structural colors were successfully obtained

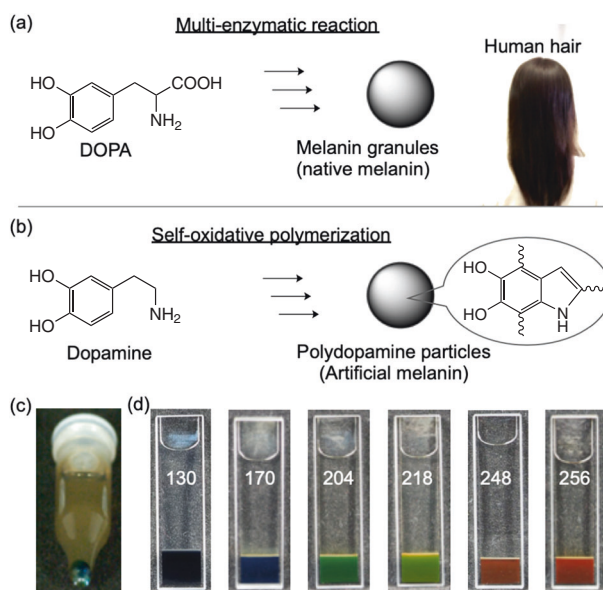


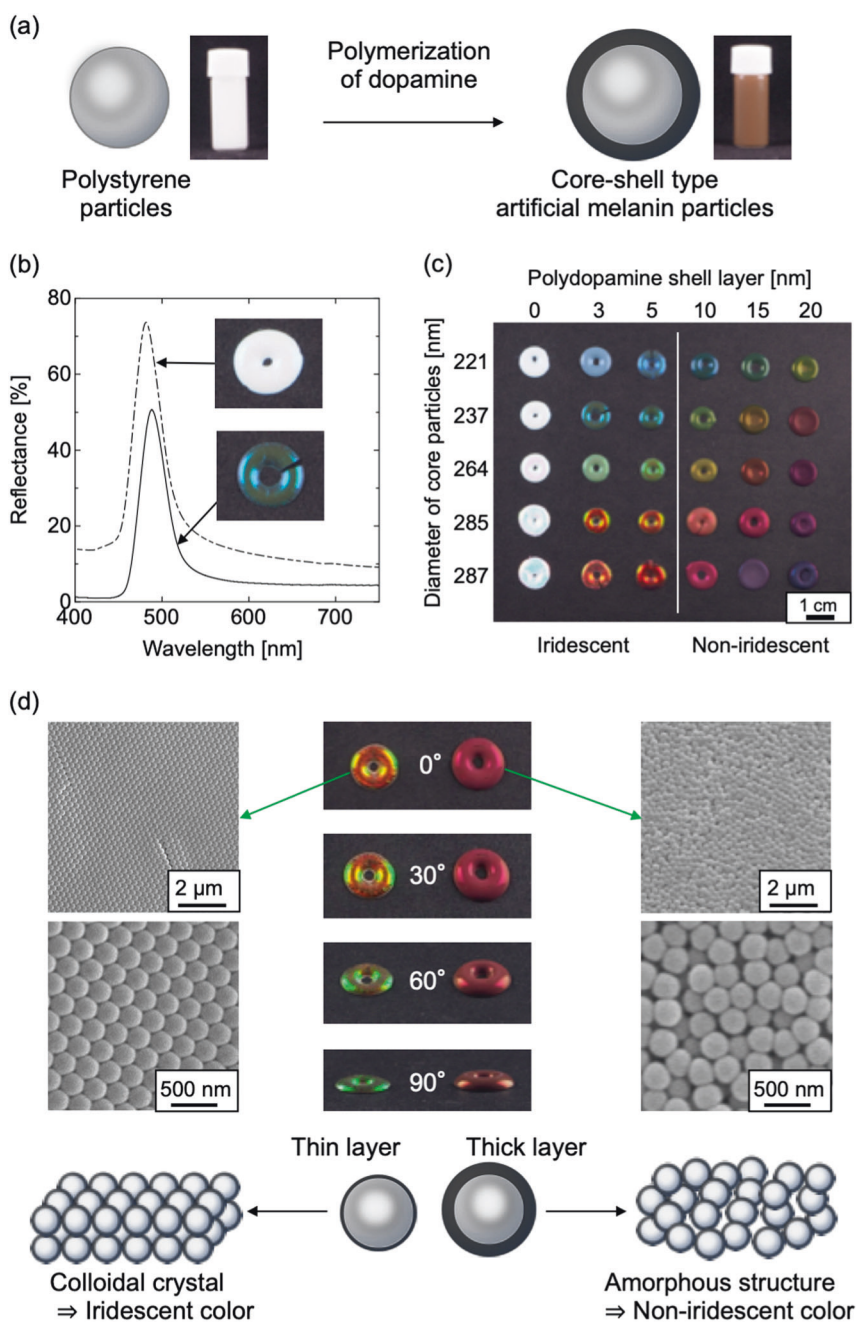
Fig. 2 Schematic illustration of **a** melanin granules and **b** polydopamine particles that mimic melanin granules. **c** Green structural colors discovered early in the experiment. **d** Structural colors from the assembly of uniform-sized polydopamine particles in water (particle concentration: 50 wt%). Numbers represent the diameters of the polydopamine particles (in nm). Adapted with permission from ref. [39]. Copyright 2015 the Royal Society of Chemistry

using polydopamine particles with diameters of 130–256 nm, indicating that the colors were controlled by the diameter of the polydopamine particles (Fig. 2d) [39]. Shortly after our first paper, Xiao et al. reported the occurrence of structural coloration based on thin-film interference from a layer created by the assembly of polydopamine particles [40]. In this case, the changes in structural color were controlled by the thickness of the film using the hygroscopicity of the polydopamine particles [41].

To produce bright structural colors via the assembly of colloidal particles, it is necessary to add black materials, such as carbon black [42–44], cuttlefish inks [45], and polypyrrole [46], for enhanced color saturation. Because polydopamine particles, a mimetic material of melanin, are a component of the structural color material and absorb scattered light, bright structural colors were obtained without black additives. Structural colors from polydopamine particles were seen under strong illumination. However, the particles had dark coloration under normal illumination, such as in a room, due to excess absorption of light by the polydopamine. While this property is useful for anticounterfeit applications as reported by Cho et al. [47], this property has disadvantages in terms of use for other applications, e.g., printing inks, colored materials, and reflection-type devices.

In the structural coloration of bird feathers, there is a case in which a structural color with high visibility is obtained by decreasing the absorbance of melanin through the alteration of its hierarchical structure. Turkey feathers present

Fig. 3 **a** Design of core–shell type artificial melanin particles. **b** Reflection spectra of structural color pellets from polystyrene core particles (dotted line) and core–shell type artificial melanin particles (solid line). Insets show photographs of the structural color pellets. **c** Structural color pellets prepared from artificial melanin particles with different core diameters and polydopamine shell layer thicknesses. **d** Scanning electron microscope (SEM) images and photographs taken from different viewing angles of structural color pellets composed of artificial melanin particles. Adapted with permission from ref. [49]. Copyright 2016 Nature Publishing Group



iridescent structural colors from the assembly of hollow-shaped melanin granules [48]. Inspired by this finding, we designed core–shell type particles, called artificial melanin particles in this paper, as shown in Fig. 3a [49]. Core–shell particles composed of monodispersed polystyrene particles and a polydopamine shell layer were obtained by polymerization of dopamine in the presence of core materials. The blackness of the particles could be controlled through varying the thickness of a polydopamine shell layer, which was dependent on the polymerization conditions.

Artificial melamine particles suspended in water were dropped onto a silicone rubber plate, and the suspensions

were allowed to dry, producing pellet samples. While the polystyrene core particles formed milky white colored pellets with iridescent colors, high visibility structural color pellets were obtained from the artificial melanin particles [49]. Figure 3b shows the reflection spectra of the pellet surfaces made from polystyrene and artificial melanin particles. The intensity of the reflectance peak at ~480 nm was higher in the pellet prepared from polystyrene particles due to the blue structural color of these particles. However, the pellet appeared white to the human eye because of the high overall reflection in the visible range due to light scattering. On the other hand, for the artificial melanin particles, the

reflection due to light scattering substantially decreased due to the broad-spectrum light absorptivity of the polydopamine shell layers, much like the properties of melanin. Although the reflectance for structural color was also reduced, the visibility of colors to the human eye dramatically improved. By controlling the size of the core particles and the thickness of the polydopamine shell layer of the artificial melanin particles, nearly the full range of structural colors could be obtained (Fig. 3c) [49].

Particle arrangements and angular dependence of structural colors

The periodic arrangement of melanin granules in peacock feathers produces an iridescent structural color in which the hue of the color changes with the viewing angle [17]. On the other hand, the feathers of *Cotinga maynana* show a non-iridescent blue structural color independent of the viewing angle [50]. Inside the feathers of *Cotinga maynana*, an amorphous structure consisting of a nanostructured keratin-air matrix is formed. In amorphous structures with only short-range order, non-iridescent structural colors caused by wavelength-specific reflection appear [51, 52]. Control of the spectral angular dependence of structural coloration is an important subject for practical applications. Interestingly, in the structural color pellets produced by the assembly of the artificial melanin particles, the angular dependence of the structural colors was easily controlled by the thickness of the polydopamine shell layers (Fig. 3d) [50]. Core-shell particles with thin polydopamine shell layers (<5 nm) formed colloidal crystal structures, and angle-dependent structural colors appeared. In contrast, particles with a thick shell layer (more than 10 nm) showed angle-independent structural colors due to their amorphous structure. These differences are believed to be due to the surface roughness of the particles.

Particle mixing can also control the angular dependence of structural coloration. When artificial melanin particles with different sizes were mixed to form pellets, the particle arrangement changed in accordance with the mixing ratio, and the angular dependencies of the resulting colors changed [53]. In this case, the average size of the particles, which affects the structural coloration, can be adjusted stepwise by particle mixing. Thus, neutral structural colors can be obtained by simple particle mixing [53].

Effects of components of artificial melanin on structural coloration

Melanin is known as a black component of human hair. Individuals differ in hair color because of differences in

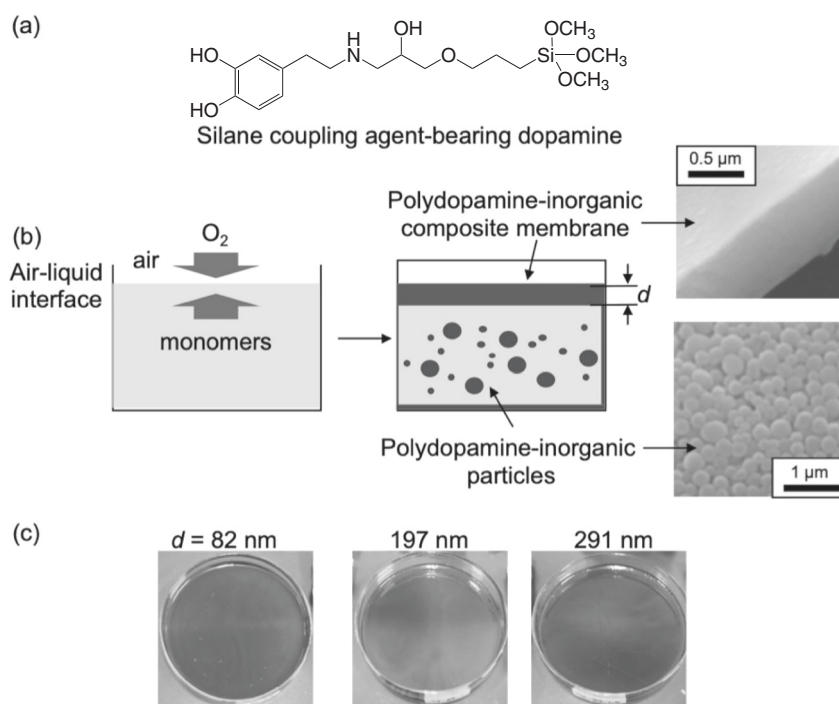
melanin content and composition. Thus, the effect of the melanin precursor on the structural coloration was investigated. In addition to dopamine, artificial melanin particles were prepared from DOPA and norepinephrine, which are melanin precursors similar to dopamine in vivo [18]. The polymerization of DOPA and norepinephrine on the surface of polystyrene particles was also successfully performed by adding an oxidant, i.e., sodium periodate [54]. PolyDOPA-coated particles had the same tendency as polydopamine-coated particles; rough surface core-shell particles were obtained with the formation of non-iridescent structural color pellets. In contrast, polynorepinephrine produced a smooth and thick shell layer compared with that of polydopamine, and pellets consisting of the particles had iridescent structural coloration. The difference in the shell surface morphologies, which affects the structural coloration, is probably due to differences in polymerization mechanisms [55–57]. The results indicated that the angular dependence of the structural coloration can be controlled by the molecular composition of the artificial melanin particles.

Copolymerization of dopamine and functionalized dopamine also provides interesting structural color materials. When copolymerization of dopamine and silane coupling agent-bearing dopamine was performed under static conditions, a polydopamine-inorganic composite membrane was formed at the air-liquid interface (Fig. 4a, b) [58]. As a result of the copolymerization of both organic and inorganic components, a robust and thick membrane of ~100–300 nm was obtained, possessing structural coloration due to thin-film interference. In this case, polydisperse polydopamine particles, which were simultaneously generated in the lower portion of the membrane, effectively suppressed light scattering, producing high visibility coloration (Fig. 4b, c) [58]. Prior to our report, Wu et al. reported that similar coloration could be obtained by transferring the hierarchical structure of polydopamine films and particles onto a substrate [59]. These structural colorations are similar to those of pigeon feathers, which are derived from a hierarchical structure consisting of keratin membranes and melanin granules [60]. Recently, Zhang et al. showed that structural colors can also be obtained by forming a thin film with a polydopamine coating on a substrate without using polydopamine particles [61].

Varying structural colors depending on particle shapes

While solid spherical particles are commonly used as components of artificial structural colors produced by the assembly of colloidal particles, various shapes of particles can produce structural coloration in bird feathers. In the structural colors of peacock and turkey feathers, rod-like melanin granules [17] and hollow melanin particles [48] are components, respectively,

Fig. 4 **a** Chemical structure of silane coupling reagent-bearing dopamine. **b** Schematic illustration of the synthetic process generating the hierarchically structured membrane and particles. Insets show the SEM images of the membrane and particles. **c** Photograph of the structural color obtained. Adapted with permission from ref. [58]. Copyright 2017 The Society of Polymer Science



leading to speculation that particle shape is an important factor in the determination of structural coloration.

Ellipsoidal particles with different aspect ratios were prepared by stretching a polymer film containing spherical artificial melanin particles [62]. The reflection wavelength of the pellets assembled from ellipsoidal particles blueshifted as the aspect ratio of the particles increased (Fig. 5) [62]. Because the ellipsoidal artificial melanin particles were mostly horizontally oriented in the pellet, the reflection wavelength was blueshifted depending on the minor axis length of the ellipsoidal particles. As the solution in which the particles were dispersed dried, the ellipsoidal particles assembled in a different manner than the spherical particles. While the spherical particles assembled at the edge of the droplet and dried to form donut-shaped pellets, flat-shaped pellets were formed from the ellipsoidal particles that tended to be adsorbed near the air-water interface.

To produce robust hollow particles, core-shell particles with a smooth and thick shell layer were prepared by repeating the polydopamine coating process [54]. Then, the polystyrene core particles were dissolved by treatment with tetrahydrofuran, producing hollow artificial melanin particles. While the arrangement of the hollow particles in the pellet samples was disordered compared with the arrangement of the core-shell particles due to partial collapse, bright structural color was obtained from the assembly of the hollow artificial melanin particles. The structural coloration of the pellets from hollow particles

was blueshifted compared with that of the core-shell particles due to the difference between the refractive indices of the two types of particles [54]. The preparation of melanin materials that maintain a hollow structure was reported by Yi et al. [63]. After curing the assembled structure of the polydopamine-coated polystyrene particles with a silane coupling agent [64, 65], a reverse opal structure was obtained by removing the polystyrene core. These materials are expected to be used in sensors because the structural colors change according to the refractive index of the solvent penetrating the core of the hollow particles. The curing of photonic structures is an important issue. Techniques have been reported for curing the assembly structure of particles using polyurethane particles [66] and alkali vapor treatment [67].

Hairy artificial melanin particles were obtained by preparing polymer brush-grafted particles, using surface-initiated living polymerization [68]. Atom-transfer radical polymerization (ATRP) initiating groups were selectively introduced on the surface of the polydopamine particles by adding ATRP initiator-bearing dopamine in the later stages of polydopamine particle preparation. After grafting the polymer chains, the hairy polydopamine particles maintained monodispersity. The structural coloration of the hairy artificial melanin particles depended on the length and type of the polymer chains. In the future, control of structural coloration in response to various stimuli is expected to be achieved by the application of stimulus-responsive polymers.

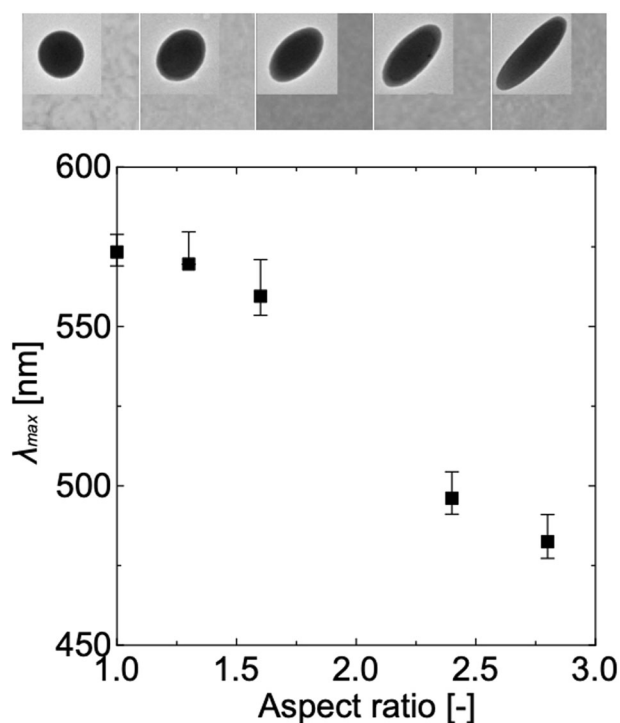


Fig. 5 Maximum value (λ_{max}) of the reflection spectrum of structurally colored pellets prepared from particles with various aspect ratios. Above, digital microscope images of color pellets with inset transmission electron microscope (TEM) images of particles. Adapted with permission from ref. [62]. Copyright 2019 American Chemical Society

Development of structural color inks

In the development of structural color-based inks, the thickness of the ink film is an important consideration. Film thickness-controlled structural color films were prepared by dip coating artificial melanin particles [69]. A close-packed structure was formed by pulling at low speed (0.1 $\mu\text{m/s}$), and uniform and bright structural color films were obtained. The film thickness and reflectance were controlled by the particle concentration during dip coating. When the assembly of artificial melanin particles formed a film $\sim 10 \mu\text{m}$ in thickness, the artificial melanin particles effectively absorbed both transmitted and scattered light, appearing brightly colored to the human eye. Furthermore, the films obtained had bright structural colors independent of the background, including black and white colors [69]. The surface properties of the substrates affected the adhesion of the artificial melanin particles and their structural coloration [70].

Structural color printing was demonstrated using the ink-jet method with water-based inks containing artificial melanin particles, as artificial melanin particles are dispersed well in water due to a high zeta potential of approximately -50 mV [49]. After ink-jet printing the particles, a dome-shaped structure sized $\sim 100 \mu\text{m}$ was

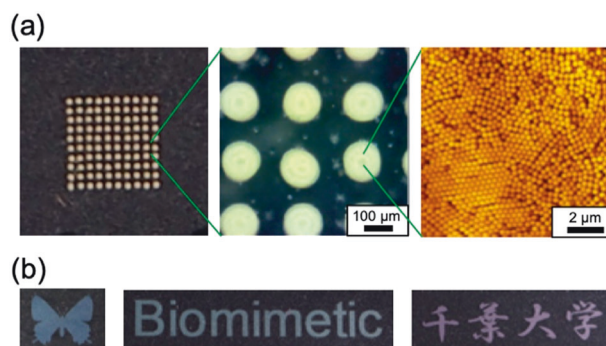


Fig. 6 **a** Digital microscope and atomic force microscope (AFM) images of structural color prints produced by the ink-jet method. **b** Structural colors of artificial melanin particles obtained by the ink-jet method. Diameters of the artificial melanin particles are blue: 221 nm, green: 274 nm, and red: 292 nm

obtained (Fig. 6a). Structural color was observed in the dome-shaped structure formed by the assembly of artificial melanin particles (Fig. 6a). Depending on the size of the particles used, the color of the ink could be controlled (Fig. 6b). By coating the surface of the polydopamine particles with surfactants, the dispersibility not only in water but also in nonpolar solvents such as toluene, heptane, and hexane could be improved [71]. Due to the difference in the refractive indices, the reflection wavelength of the structural colors depended on the dispersion solvent. Furthermore, the structural color could be controlled by altering the interparticle distance using magnetism in the presence of a surfactant with holmium, which is a lanthanide element. Our research group has undertaken the development of holmium composite particles that respond more quickly to magnetism for magnetic field-assisted structural color tuning [72].

Due to the high dispersibility of artificial melanin particles in water, W/O emulsions containing particles were easily obtained using membrane emulsification (Fig. 7a) [73]. Then, the W/O emulsions were heated, and the process of evaporation was observed by microscopic measurements. The emulsions were a brown color that is typical of polydopamine due to light absorption and gradually decreased in diameter. After the water completely evaporated, spherical photonic materials with a diameter of $10 \mu\text{m}$ were obtained, exhibiting structural coloration even when dispersed in a solvent. Continuous production of W/O emulsions using a microdevice has produced fibrous photonic materials (Fig. 7b) [73]. The resulting fibers had an angle-independent structural color. Recent work by Xiao et al. demonstrated the possibility of preparing spherical photonic materials using a simple one-pot reverse emulsion process [74]. These materials will be useful in the development of dye-free coloration technology for creating various three-dimensional colloidal architectures.

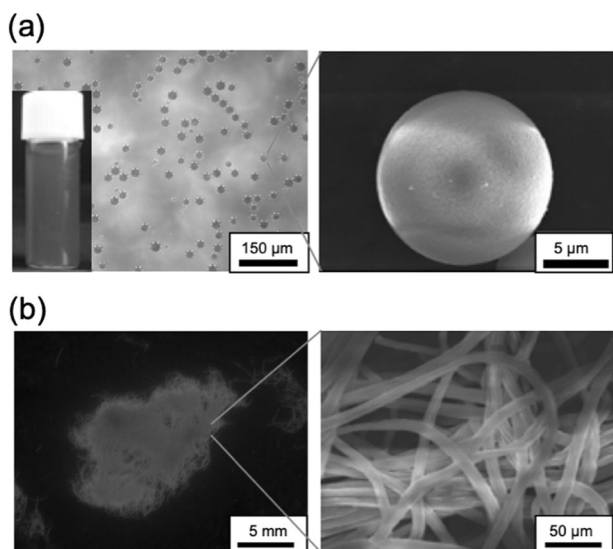


Fig. 7 Photographs and SEM images of **a** spherical photonic materials and **b** fibrous photonic materials from artificial melanin particles. Adapted with permission from ref. [73]. Copyright 2018 American Chemical Society

Summary and perspective

Polydopamine-based artificial melanin particles, inspired by natural melanin and its important role in structural coloration, were used to produce bright structural colors by controlling light reflection and absorption. A variety of structural coloration characteristics have become possible through our investigations of the assembly structure, composition, shape, and application of artificial melanin particles. As polydopamine is highly biocompatible, in addition to the development of structural color-based ink, applications are expected to be found in various fields, e.g., cosmetics that touch the skin. While melanin has been shown to be important for structural coloration, the role of melanin in vivo and the mechanism by which periodic structures composed of melanin are generated are still relatively unknown. Polydopamine is a material that mimics natural melanin in terms of both composition and structure and is easily synthesized with periodic structures. This simple and novel process using polydopamine-based artificial melanin particles will be useful in basic research on structural colors in nature and in practical applications.

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Compliance with ethical standards

Conflict of interest The author declares he has have no conflict of interest.

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