

## Flexible, Crack-Free, and Robust Structural Color Films Composed of Melanin and Keratin

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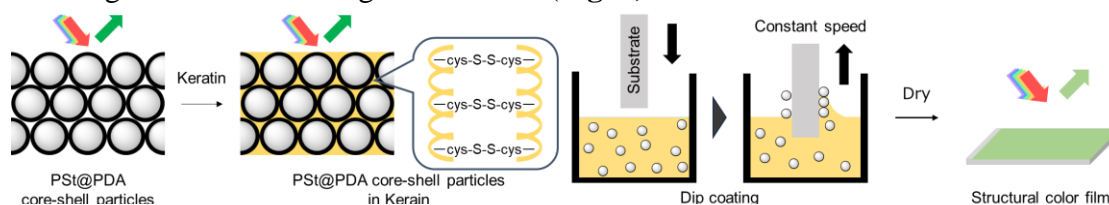
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### Abstract

In peacock feathers, melanin granules are regularly arranged, and a photonic crystal structure that generates beautiful structural colors exists. While there have been reports on the fabrication of structural color materials using artificial melanin particles, improving the mechanical strength of these materials is a crucial challenge for their practical application. This study focuses on the fact that peacock feathers are composed of melanin and robust keratin, and by filling the gaps between particles with keratin, we created structural color materials with excellent mechanical strength. The resulting structural color films are highly durable and crack-free, suggesting that this method improves the mechanical strength of structural color materials.

### 1. Introduction

In nature, many organisms exhibit vivid structural colors. For example, in peacock feathers, submicron-sized melanin granules are arranged in a regular pattern, forming a photonic crystal structure that generates structural color. Inspired by the vivid structural coloration of peacock feathers, we developed bright structural color materials using artificial melanin particles composed of a polystyrene (PSt) core and a polydopamine (PDA) shell that mimics melanin, referred to as PSt@PDA particles.<sup>1,2)</sup> However, structural color materials based on PSt@PDA particles, although widely studied, suffer from low mechanical strength and are prone to cracking.<sup>3)</sup> In contrast, biological organisms in nature that exhibit structural coloration are generally crack-free, many of which are composed primarily of keratin.<sup>4)</sup> Keratin is a multifunctional biopolymer with excellent robustness and elasticity, and it plays a critical role in a variety of biological functions. Due to these properties, keratin-based materials are widely applied in bioinspired designs, biomedical fields, and environmentally sustainable fiber-reinforced composites.<sup>5)</sup> In this study, crack-free structural color materials with superior mechanical strength were fabricated. A keratin solution was added to a dispersion of PSt@PDA particles, and upon drying, the keratin matrix filled the voids between the particles, effectively immobilizing them. Additionally, structural color films were produced using a dip-coating method, and the color characteristics and physical properties of the resulting films were investigated in detail (**Fig. 1**).



**Fig. 1** Preparation of PSt@PDA core-shell particles in keratin and schematic diagram of the preparation of the structural color film by the dip coating method.

## 2. Experiment (or Theory)

The PSt@PDA core-shell particles were synthesized by polymerizing dopamine in the presence of PSt core particles. Subsequently, a water-soluble keratin solution was added to the dispersion of PSt@PDA particles. The resulting particle dispersion was dried to prepare pellet samples, and the effect of keratin on the structural coloration of the pellets was investigated. Finally, structural color films were fabricated using a dip-coating method, and the color characteristics and physical properties of the films were studied in detail.

## 3. Results and discussion

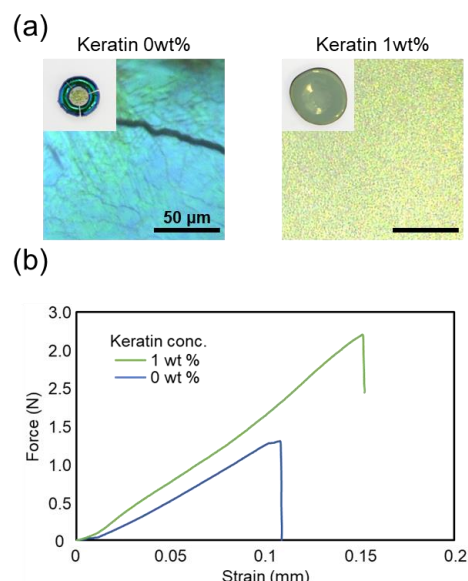
Pellet materials were prepared both before and after the addition of keratin. Optical microscopy observations of the pellet surfaces revealed that they were crack-free (Fig. 2(a)). This indicates that the addition of keratin contributed to stress relaxation through an increase in viscosity and a reduction in surface tension, which in turn influenced the particle aggregation behavior. To evaluate the mechanical strength of the fabricated pellet materials, a simple puncture test was conducted using a compression tester and a puncture probe to assess the strength of the films and materials (Fig. 2(b)). It was confirmed that the mechanical strength of the pellets with added keratin had increased. Furthermore, structural color films were fabricated using the dip-coating method. To improve flexibility, the films were coated onto PET substrates. Unlike the films made from conventional melanin particles, which were easily delaminated, the films prepared in this study showed no signs of delamination, demonstrating improved flexibility and robustness. Additionally, a bending test was performed on the films using a small-scale tabletop durability tester. The results confirmed that even after 100,000 bending cycles, the structural integrity and structural coloration of the films were maintained without degradation (Fig. 3).

## 4. Conclusions (or Summary)

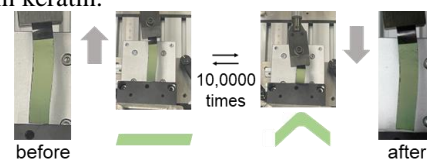
In this study, we successfully prepared flexible structural color films with excellent mechanical strength and no cracks by adding water-soluble keratin to a melanin particle dispersion. The method proposed here enables simpler material fabrication and expands the potential of structural color materials.

## References

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**Fig. 2.** (a) Optical microscopic images and photographs of pellet samples prepared from PSt@PDA particles and PSt@PDA particles containing keratin. (b) Testing force as a function of the strain for the structural color pellets composed of PSt@PDA and PSt@PDA in keratin.



**Fig. 3.** Photographs of the structural color film after 10,000 bends.