



Simple fabrication of colloidal crystal structural color films with good mechanical stability and high hydrophobicity



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ABSTRACT

Weak mechanical stability and low hydrophobicity have been considered as major obstacles for actual applications of structural color films from colloidal crystals. This paper presents these films with good mechanical properties and high hydrophobicity by employing a simple preparation strategy. With this strategy, the mechanical property of the structural color films was remarkably improved by the structure lock effect of polyacrylate adhesive when its latex was used as a co-assembly component with polystyrene microspheres. Likewise, the hydrophobicity of structural color films was enhanced by using cationic surfactant. Furthermore, the obtained films exhibited several unique characteristics, such as good mechanical stability, high hydrophobicity, and bright structural color. Therefore, the combination of hydrophobicity and mechanical stability was significant for potential architectural applications of paint and external decoration.

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1. Introduction

Monodisperse colloidal particles undergoing self-assembly into an ordered array structure provides a simple and convenient approach to prepare colloidal crystals [1–4], which can control light propagation caused by photonic stop band [5–9]. In particular, the photonic stop band in the visible region is of special interest because this band can directly convert light to color (structural color), that is potentially useful for display devices [10,11], colorimetric sensors [12,13], optical filters, paints, and photonic paper [14–17]. However, colloidal crystals exhibit very low durability because of weak interaction among colloidal particles [18]. Furthermore, the structures of colloidal particles do not possess high hydrophobicity because of charge characteristics [19]. The persistence and the hydrophobicity of the structural color films from colloidal crystals are crucial for actual applications in full-color displays, paints, and photonic paper. Once the periodic array is destroyed, structural color likely disappears [20]. At the same time, these applications may be limited by pollution caused by dust particles, snow, and fog, among others. Therefore, research on structural color films with both mechanical stability and hydrophobicity is of practical significance.

Scientists developed effective methods to improve the persistence of colloidal crystal [21]. For example, Finlayson [22] synthesized polymer spheres with core–shell structure. Cross-linked polystyrene sphere is used as a core and soft polyethylacrylate is used as a shell. Colloidal crystal film with tough mechanical strength is subjected to a roller process, producing 100 μm thick opal films via squeeze/linear shear. Song [23,24] obtained colloidal crystal film with tough mechanical strength by vertical deposition of polymer latex spheres with a hard polystyrene core and an elastometric poly(methyl methacrylate–acrylic acid) shell. Fudouzi [25] prepared a silicone rubber structural color sheet with good mechanical properties by embedding polymer spheres into a silicone elastomer sheet.

In this article, we have developed one more simple approach to fabricate colloidal crystal structural color films with robust mechanical properties and high hydrophobicity from PS microspheres and polyacrylate (PA) by using a thermal-assisted self-assembly method. Compared with other methods, this new approach shows the following advantages: (i) PS microspheres were easily obtained by classical emulsion-free polymerization [26,27]; (ii) fabrication was very simple because the mixture of PS and PA latex can be used to assemble structural color films directly; (iii) the obtained colloidal crystal structural color films exhibited good mechanical properties through the structure lock effect of polyacrylate adhesive; and (iv) structural color films were easily changed into a

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hydrophobic structure. These advantages are essential for actual architectural applications in paint and external decoration.

2. Experimental

2.1. Materials

Styrene (St) and sodium dodecyl sulfate (SDS) were purchased from Damao Chemical Reagent Factory (Tianjin, P.R. China). Methyl methacrylate (MMA), butyl acrylate (BA), and potassium peroxydisulfate were purchased from Bodi Chemical Industry Co., Ltd. (Tianjin, P.R. China). Cetyltrimethylammonium chloride was purchased from Fuchen Chemical Reagent Factory (Tianjin, P.R. China). Styrene was processed in accordance with a previously described method [19]. Other reagents were used without further purification.

2.2. Preparation of monodispersed PS suspension

Monodispersed PS spheres with different sizes were prepared by emulsifier-free emulsion polymerization performed as follows: 100 mL of deionized water, an appropriate amount of sodium dodecyl sulfate with lower than the critical micelle concentration (CMC), and 10.0 g of styrene were placed in a 250 mL four-necked flask equipped with a nitrogen bubbler, a mechanical stirrer, a temperature indicator, and a reflux condenser. The system was stirred at 250 r/min and maintained at 85 °C by using a water bath under N₂. Then, 0.100 g of potassium peroxydisulfate was added; polymerization was carried out at 85 °C for 5 h with nitrogen protection. PS latex was formed and stored for the subsequent self-assembly experiment.

2.3. Preparation of PA adhesive emulsion

Approximately 100 mL of deionized water and 0.6 g of sodium dodecyl sulfate were placed in a 250 mL four-necked flask equipped with a nitrogen bubbler, a mechanical stirrer, a temperature indicator, and a reflux condenser. The system was stirred at 250 r/min for 20 min. Afterward, 5.0 g of MMA and 5.0 g of BA were added and the mixture was maintained at 85 °C by using a water bath under N₂. Then, 0.100 g potassium peroxydisulfate was added, and polymerization was carried out at 85 °C for 5 h with nitrogen protection. Polyacrylate (PA) adhesive latex was formed and stored for subsequent self-assembly experiment.

2.4. Preparation of the robust crystal films with high hydrophobicity

The robust films are immersed into 0.01 g/mL cetyltrimethylammonium chloride solution for 28 h. The long carbon chain bearing a positive charge can be absorbed onto the surface of the PS/PA structural color films, and the robust crystal films with high hydrophobicity are obtained.

2.5. Characterization

The Z-average hydrodynamic diameters of the particles were determined by using a particle size analyzer (Zetasizer 1000, Malvern, UK). Scanning electron microscopy (SEM) images were obtained using a Nova NanoSEM 450 scanning electron microscope to observe the surface morphological characteristics of the films. All of the samples were coated with gold before these samples were observed. Water contact angle (CA) measurements were conducted using a Phoenix-300 contact-angle system (SurfaceTech, Korea). Surface topographical images were obtained by using an atomic

force microscope (AFM; Nanoscope IIIA+, Veeco Instruments, USA) to perform topographical characterization of the films.

The reflection spectra of the films were obtained at a scan speed of 300 nm/min by using a HITACHI U-4100 spectrophotometer with a slit width of 8.00 nm. Optical photos of the structural color films before and after modification were captured with a Nikon digital camera. Glass transition temperature (T_g) was measured by using a 910S DSC thermal analyzer (TA Instruments, America) at a heating rate of 5 °C/min and an N₂ flow rate of 30 mL/min. The samples were evaluated in a sealed aluminum pan with a mass of approximately 10.0 mg.

3. Results and discussion

3.1. Preparation and characterization of emulsion

The concentration of emulsifier was a key factor in emulsion polymerization. The effects of sodium dodecyl sulfate on sizes and polydispersity index (PDI) of PS microspheres are summarized in Table 1. For the PS latexes (samples a-d), hydrodynamic particle sizes of the microspheres can be easily controlled by changing the concentration of sodium dodecyl sulfate, which was necessary to obtain different structural colors. After the concentration of SDS was increased from 0.032wt% to 0.054wt% of the emulsion weight, four kinds of microspheres (hydrodynamic particle sizes of 301, 288, 255, and 223 nm) were obtained. The actual diameters were 265, 251, 233, and 190 nm, respectively, which were measured by using SEM.

At the same time, PDI was <0.1 and zeta potential was <−30 mV, indicating that the emulsion was monodispersed and stable [22]. Polyacrylate (PA) adhesive emulsion was formed by the emulsion polymerization, and the hydrodynamic particle size was 42 nm, PDI was 0.08, zeta potential was about −86 mV, indicating that the PA particles were very stable in the latex system.

3.2. Fabrication and structure of the polymer crystal films

Scheme 1 illustrates the fabrication procedure of hydrophobic PS/PA structural color films. PS and PA emulsions were mixed (15:1, v:v), and the mixture was then deposited dropwise onto a glass substrate. The substrate was placed on a hot plate at 85 °C to assist self-assembly. After thermal-assisted self-assembly was completed, the ordered structure of PS particles was locked by PA due to PA's three advantages. Firstly, the small PA nanoparticles can enter the voids among PS particles instead of air; secondly, PA (T_g, −21 °C) would melt when temperature rise to 85 °C; finally, the good adhesion of PA can increase the interaction between PS nanoparticles. Then, these mechanically stable films can be further treated. The zeta potential of PS is <−30 mV (Table 1.). Thus, the long carbon chain bearing a positive charge can be absorbed onto the surface of the PS/PA structural color films. The wettability of tough structural color films could be altered to hydrophobicity from hydrophilicity.

The high quantities of polymer photonic crystal structural color films were obtained when the volume ratio of PS suspension to PA emulsion was 15:1(v:v) (Fig. S1) by using the thermal-assisted self-assembly method published in our previous studies [19,20]. The SEM images of PS, PS/PA, and modified PS/PA films are shown in Fig. 1. The diameters shown in Fig. 1 a–d were 265, 251, 233, and 190 nm, respectively.

Fig. 1 a–d are different from a'–d' and a''–d''. The voids between PS particles in an orderly array were filled by air when PS spheres assembled individually. By contrast, the voids between PS particles in an orderly array were filled with PA (Fig. 1 a'–d' and a''–d'') when PS spheres and PA co-assembled. PA with small particle sizes

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