

## Supporting Information

### **Liquid-immune structural colors with angle-independence inspired from hollow melanosome**

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### **Experimental**

#### **1. Materials**

Styrene (St), methacrylic acid (MAA), ammonium persulfate (APS) and polydimethylsiloxane (PDMS) were purchased from Damao Chemical Reagent Factory (China). Dopamine hydrochloride and Tris(hydroxymethyl)aminomethane (Tris) were purchased from Yuanye Bio-Technology Co., Ltd (China). 3-aminopropyltriethoxysilane (APTES) was purchased from Aladdin Industrial Corporation (China). Ethanol, isopropanol, hydrochloric acid (HCl) and tetrahydrofuran (THF) were purchased from Guanghua Sci-Tech Co., Ltd (China). Glass substrates (2.4 cm × 2.4 cm) were obtained from commercial approach. Deionized water (18.2 MΩ·cm<sup>-1</sup>) was used in all procedures. Prior to use, St was purified by decompress distillation to remove inhibitors and glass slides were washed by ethanol with ultrasonication. All other reagents were used as received.

#### **2. Synthesis of polystyrene microspheres**

Polystyrene (PS) microspheres were prepared as the core particles by an emulsifier-free emulsion polymerization method. In brief, deionized water (100 g), a suitable amount of St (5-10 g) and MAA (1 g) were firstly poured into a 250 mL four-necked flask, which was furnished with a temperature gauge, a stirring rod and a condenser. APS (0.15 g) dissolved in 5 mL deionized water was transferred into the reactor after the mixture was heated to 75 °C under a stirring rate of 300 rpm. Then the reaction

lasted for 8 h with nitrogen atmosphere in a constant temperature and stirring rate of 75 °C and 300 rpm, respectively. As the reaction completed, PS powders were collected by repeated centrifugation and kept in a dryer. The size distributions and morphologies of as-prepared PS microspheres are demonstrated in Figure S1.

### **3. Preparation of polydopamine-coated PS core-shell nanoparticles**

Polydopamine-coated PS core-shell nanoparticles (PS@PDA NPs) were fabricated by the oxidation polymerization of dopamine under weak base condition (see Scheme S1). Typically, 0.1 g PS powders were dispersed in Tris-HCl buffer (50 mL, 10 mM; pH = 8.5) with sonication for 30 min. Afterwards, 50-100 mg dopamine hydrochloride (DA) was directly added into the PS dispersion and maintained mildly magnetic stirring for 20 h in ambient temperature. Lastly, the PS@PDA NPs were obtained by centrifugation and rinsed several times by deionized water.

### **4. Fabrication of PS@PDA/APTES colloidal crystal films**

Primarily, PS@PDA NPs were dispersed into deionized water with a concentration of 5 wt% assisted by ultrasonication for 1 h. The obtained PS@PDA latex was mixed with APTES aqueous solution (5 wt%) in a mass ratio of 5:1 (PS@PDA to APTES), where the APTES acted as a binder to warrant the bonding among PS@PDA NPs. Afterwards, the mixture was cast onto the glass substrate and colloidal crystal film was fabricated by maintaining the glass slide at 50 °C for 30 min.

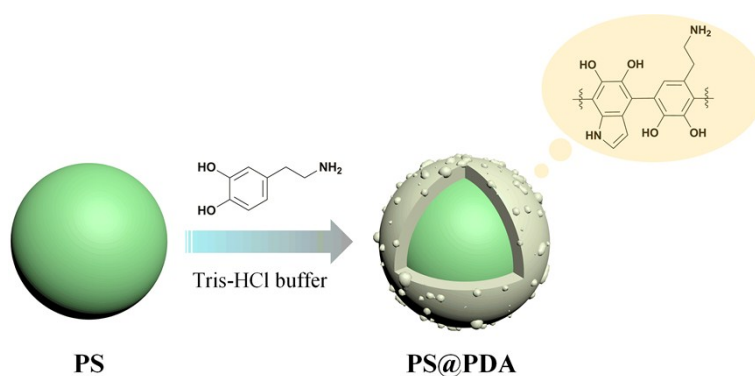
### **5. Fabrication of hollow PDA colloidal crystal films**

Hollow PDA colloidal crystal films were prepared via THF-etching for PS resolving in THF readily. Generally, as-prepared PS@PDA/APTES colloidal crystal film was carefully laid in a 25 mL beaker containing 10 mL THF. By standing for 12 h, hollow PDA colloidal crystal film was achieved.

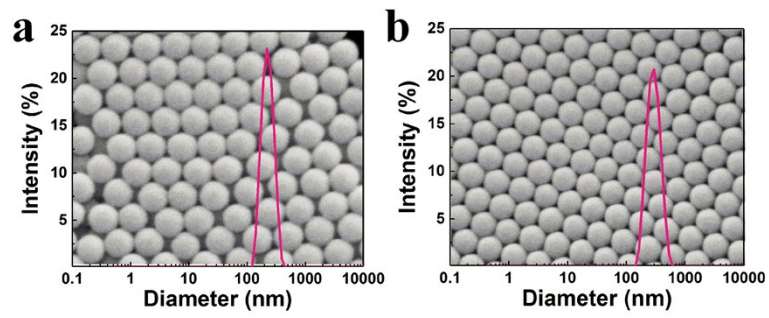
### **6. Characterization**

The surface morphologies of colloidal particles and colloidal crystal films were

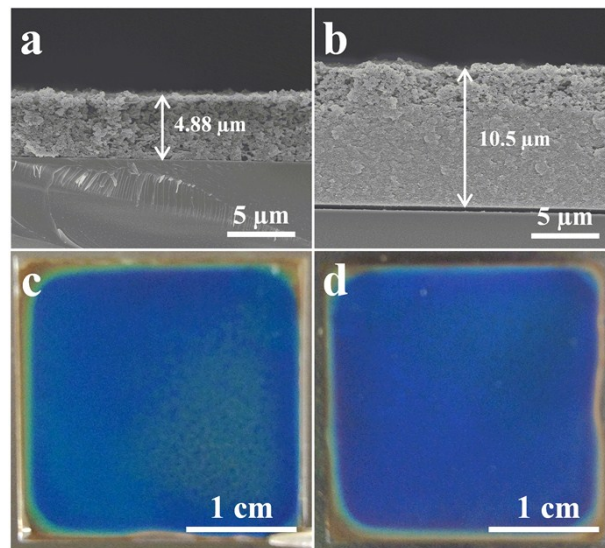
observed by field emission scanning electron microscope (FE-SEM) (Hitachi, SU-8220). Transmission electron microscope (TEM) (JEOL, JEM-2100) was applied for imaging PS@PDA NPs. Size distribution of PS microspheres was measured by dynamic light scattering (Malvern, Zetasizer Nano-ZS ZEN3600). Infrared spectra were measured with a Fourier transform infrared (FT-IR) spectroscopy (Bruker, Tensor-27). X-ray energy dispersive spectrum (EDS) was recorded on a Horiba X-Max energy dispersive spectrometer. Reflectance spectra of the films were measured at a spectrophotometer (Hitachi, U-3010). All the optical images were taken by a digital camera.



**Scheme S1.** Schematic illustration of fabricating PS@PDA NPs.



**Fig. S1.** Size distributions and morphologies of PS microspheres with mean diameters of (a) 186 nm and (b) 252 nm, respectively.



**Fig. S2** Cross-section SEM images of hollow PDA amorphous colloidal structures with the thicknesses of (a) 4.88  $\mu\text{m}$  and (b) 10.5 $\mu\text{m}$ ; (c, d) Corresponding optical images of (a) and (b), respectively.

**Table S1.** The diameters of PS microspheres, the amounts of DA, the sizes of PS@PDA NPs, and the thicknesses of PDA shell.

<b>PS (nm)</b>	<b>DA (mg)</b>	<b>PS@PDA (nm)</b>	<b>PDA shell thickness (nm)</b>
186	50	198	12
252	50	265	13
186	100	208	22

**Table S2.** The reflection peak values of hollow PDA colloidal crystals with different particle sizes under the air and liquid environments.

<b>Size (nm)</b>	<b>Reflection wavelength (nm)</b>				
	<b>Air</b>	<b>Water</b>	<b>Ethanol</b>	<b>Isopropanol</b>	<b>PDMS</b>
198	350	454	468	464	432
265	446	530	546	542	528