Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

Thermal-guided Interfacial Confinement to Fabricate Flexible Structural Color Composites for Durable Applications

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Part Ⅰ. Experimental Section

Part Ⅱ. Supplementary Figures

Part Ⅰ. Experimental Section

Materials.

4,4′-thiodiphenol was bought from Beijing Bailingwei Technology Co., Ltd. Pluronic F127 was provided by Sigma Aldrich. Absolute ethanol was bought from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China). Triethylamine was purchased from Tianjin Damao Chemical Reagent Factory. Formaldehyde solution (37 wt %) was bought from Liaoning Quanrui Reagent Co., Ltd. CB M-900 was provided by Shanghai Cabot Chemical Co., Ltd. Commercialized PVC plastics (thickness=0.5mm), TPU phone shell were purchased from a local supplier.

Preparation of Monodispersed TPFM emulsion.

A certain amount of 4,4′-thiodiphenol and Pluronic F127 were fully dissolved in a mixture of ethanol and water (70 mL) by magnetic stirring at room temperature for 20 min. The formaldehyde solution was proportionally added to the mixed solution under stirring, and then triethylamine (0.3 mL) was slowly injected to form a clear and transparent solution. The reaction solution was stirred for 10 min and then poured into the lining of 100 mL reactor at one time. The reaction kettle was sealed and placed in an oven that has been pre-heated to 150 °C, and the TPFM emulsion was obtained by hydrothermal reaction of 12 h at constant temperature. The emulsion of TPFMs with different particle sizes was prepared by adjusting the amount of ethanol, Pluronic F127 and monomer. (Table S1 and Figure S7, Supporting Information). [1]

Fabrication of APS Coatings by Spraying.

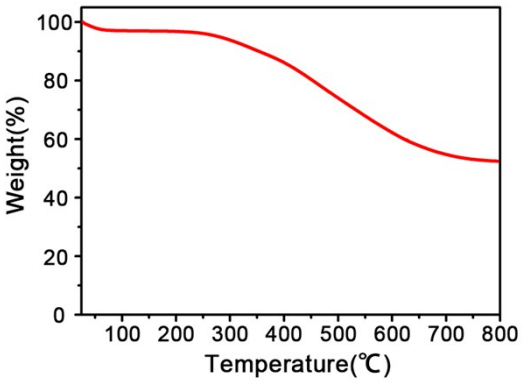
The concentration of TPFM emulsion was adjusted to 10 wt %, and a certain proportion of CB solution (0.01 g/mL) was added to form uniform emulsion through ultrasonic treatment for 1 h. The mixed emulsion was transferred into an airbrush with 0.2 mm nozzle, and then sprayed onto a thermoplastic polymer (pre-heated to 50 °C) under a pressure of 50 kPa. In our study, we adjusted the pressure and distance of the sprayer and preheated the substrate to a certain temperature, so that the microspheres were dried in the air and evenly coated on the substrate to form an APS film.

Characterization and property measurement.

SEM images were observed by a FEI Nova Nano SEM450. The average particle size was characterized by a Malvern Zetasizer NanoZS-90. Optical photos were taken by a digital camera (Nikon D7000). The reflection spectra of vertical incidence were obtained by an EQ2000 spectrometer. The scattering spectra were collected on a Hitachi U-4100 UV-vis spectrophotometer. The thermal properties of microspheres were characterized via differential scanning calorimetry and thermogravimetric analyses (TGA) (TA Instruments, USA). Samples were treated at 250 °C for 0.5 h before testing. DSC and TGA measurements were performed at the heating rate of 10 °C/min in a N_2 atmosphere. The hot pressing process was carried out in a manual forming machine QC-677T from Cometech Testing machine Co., Ltd (China Taiwan). The hot pressing time is approximately 5 s to make the films fully adhere under stable test conditions. The tensile

peel test was conducted with an AI-7000-M Tension tester from Gotech Technology Co., Ltd. The dynamic mechanical properties were measured on a DMA 1 (METTLER TOLEDO) Dynamic mechanical analyzer.

Part Ⅱ. Supplementary Figures

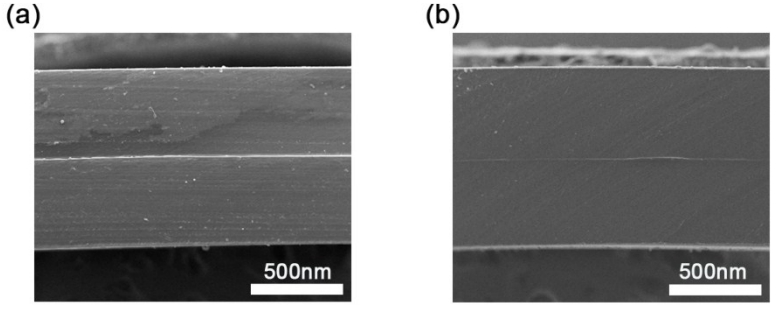


Figures S1. TGA curve of TPFMs.

Table S1. Synthesis parameters and average size of TPFMs

Sample	4,4'-thiodiphenol (g)	Formaldehyde (ml)	F-127 (g)	Ethanol (ml)	Size [a)] (nm)	PDI [b)]
TPF-1	0.25	0.25	0.03	17	283.0	0.048
TPF-2	0.225	0.225	0.025	18	239.2	0.034
TPF-3	0.225	0.225	0.05	20	197.1	0.063

[a)] All particle sizes are hydrodynamic sizes measured by Nanoparticle size measurement. [b)] PDI: Particle distribution index calculated by DLS.



Figures S2. Obvious boundaries were detected in the PVC films that were fabricated without hot pressing (a) and with hot pressing at low temperature of 50 °C (b).

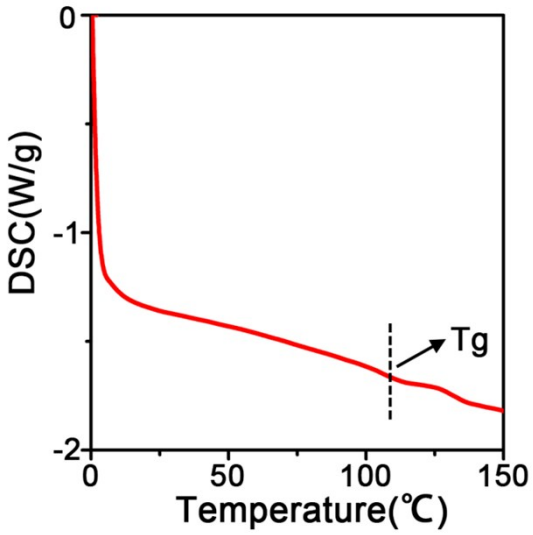


Figure S3. DSC curve of PVC.

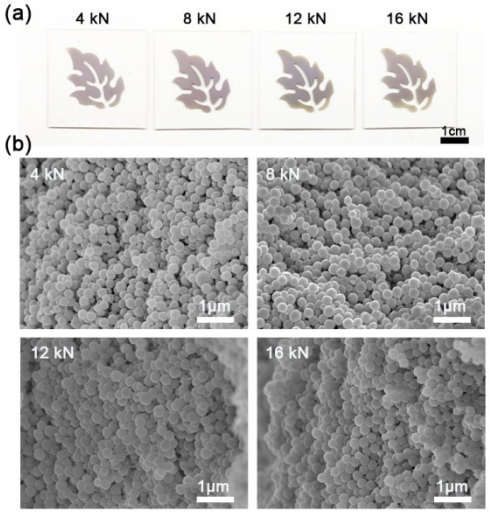


Figure S4. (a) Images of magenta-colored samples at different hot pressing pressures (4 kN–16 kN) and the temperature of 120 °C. (b) Cross-sectional SEM images of the above samples at different hot pressing pressures.

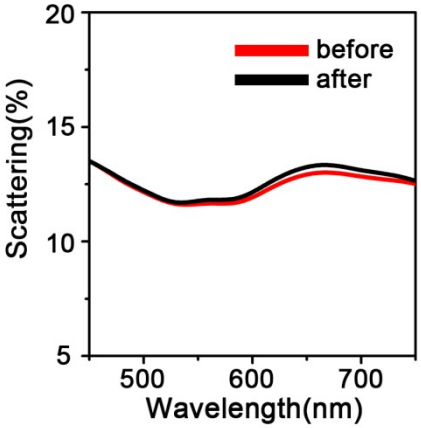


Figure S5. Scattering spectra of the micro-region at the most strained position of the composite film before and after 50 times bending test, measured by an angle-resolved microspectroscopy (Ideaoptics Instruments Ltd., China).

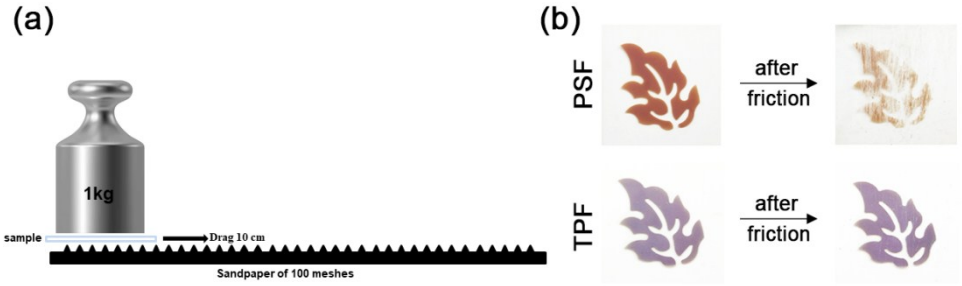


Figure S6. (a) Schematic diagram of friction experiments. Samples loaded with 1kg weight were towed 10 cm straight at a speed of 5 cm/s on 100 meshes sandpaper. (b) Comparison of PSF and TPF samples before and after 15 friction tests. The preparation of the PSF sample was conducted according to the previous literature.[2] The mixed emulsion of polysulfide (PSF) microspheres (containing 8 wt % WPU as the optimized weight ratio) was sprayed on the PVC film to form a stable APS coating. The composite film of PVC and TPFMs was prepared according to the process described in Figure 2.

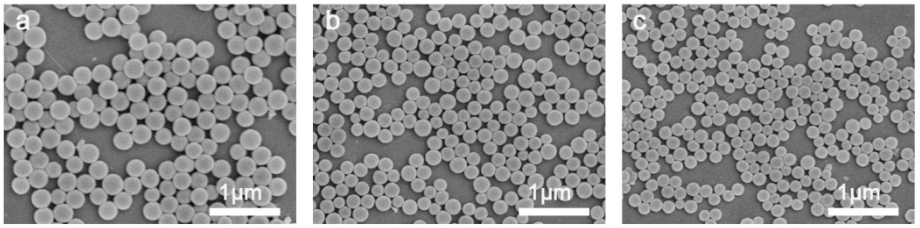


Figure S7. SEM images of TPFMs with different diameters: (a) 283 nm, (b) 239 nm, (c) 197 nm.

1 F. H. Li, B. T. Tang, S. L. Wu, W Ma and S. F. Zhang, *J. Mater. Chem. C*, 2017, **5**, 9806.

2 F. T. Meng, M. M. Umair, K. Iqbal, X. Jin, S. F. Zhang and B. T. Tang, *ACS Appl. Mater. Interfaces*, 2019, **11**, 13022.