Supporting Information

Lotus Seedpod Inspiration: Particle-Nested Double-Inverse-Opal Film with Fast and Reversible Structural Color Switching for Information Security

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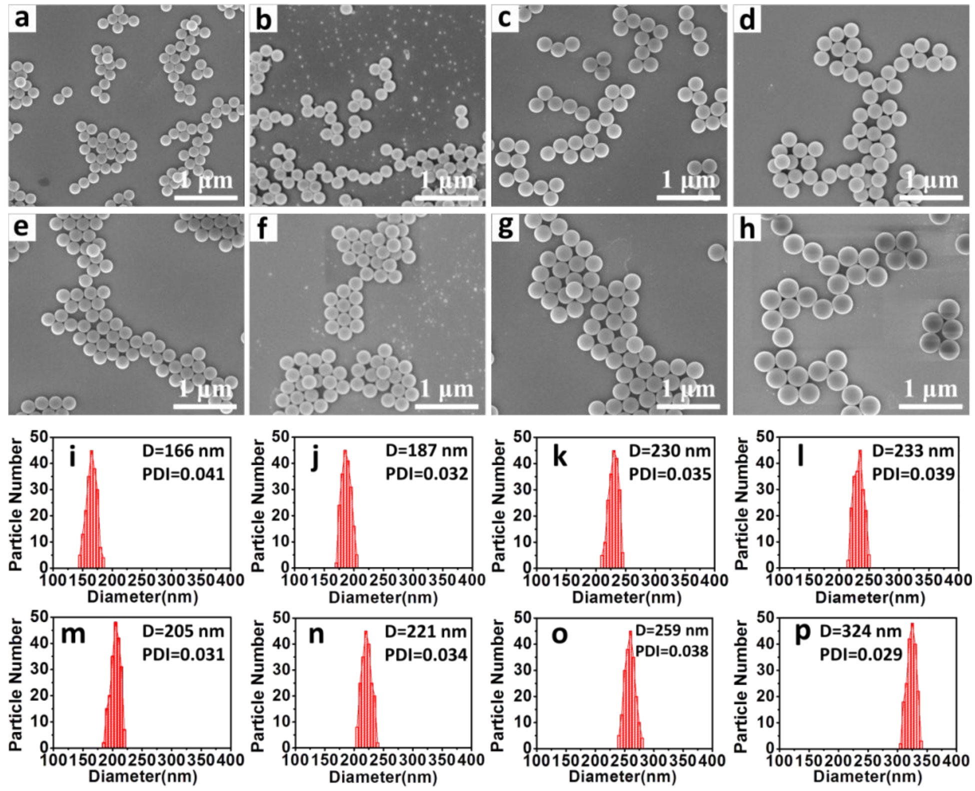

Figure S1. SEM images of the prepared PS microspheres and $PS@SiO_2$ core-shell microspheres. Particle size distribution diagrams of obtained microspheres via counting 200 nanospheres in the corresponding SEM images (a-h). The PDI and D in the figures in (i-p) represent the particle size distribution index and average particle size, respectively.

Table S1. Diameters and Zeta-potential of prepared microspheres.

Sample	Size (nm)[a]	PDI[b]	Zeta potential (mV)[c]
PS	166	0.041	-43.7
PS	187	0.032	-48.5
PS	230	0.035	-45.6
PS	233	0.039	-49.8
$PS@SiO_2$	205	0.031	-39.2
$PS@SiO_2$	221	0.034	-46.3
$PS@SiO_2$	259	0.038	-42.5
$PS@SiO_2$	324	0.029	-47.4

a: The particle diameters of prepared colloidal microspheres were obtained by counting 200 microspheres in the corresponding SEM images. b and c: Corresponding PDI and Zeta-potential of prepared microspheres were characterized by Dynamic Light Scattering.

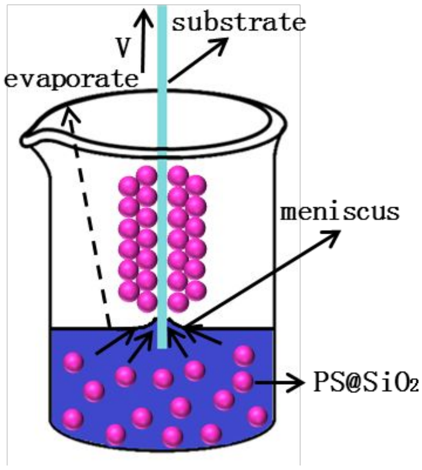

Figure S2. Schematic illustration of the dip-coating method.

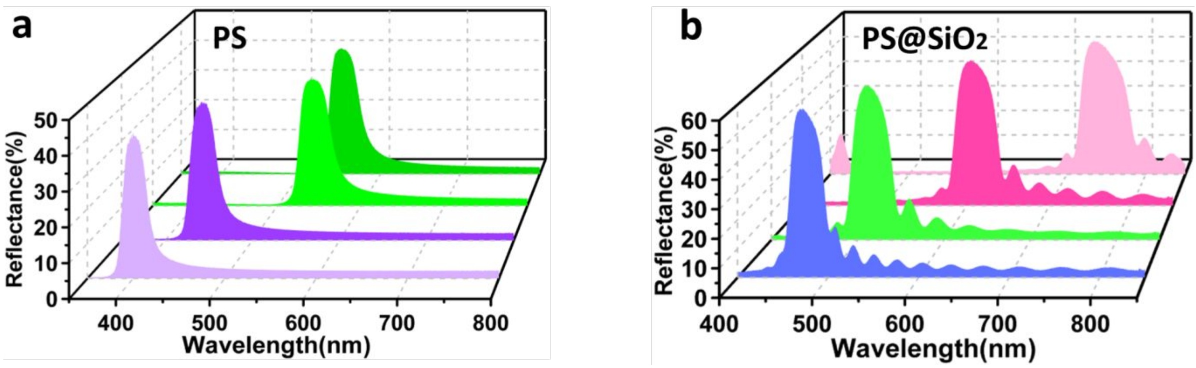

Figure S3. Reflection spectra of prepared PC templates (a, PS; b, PS@SiO_2).

Based on Bragg-Snell's law[1], the PBG of FCC structured PS PCs could be theoretically calculated as:

$$\lambda_{max} = 1.633D(n_{avg}^2 - \sin^2\theta)^{1/2} \qquad (1)$$

$$n_{avg}^2 = 0.74 \times n_{PS}^2 + 0.26 \times n_{air}^2 \qquad (2)$$

Where λ_{max} represents the maximum reflected wavelength, D is the diameter of PS microspheres, n_{avg} represents the effective refractive index, θ is the incident angle; here n_{PS} = 1.59 and n_{air} = 1.0. Through a simple formula transformation, the theoretical diameter of the prepared PS microspheres can be calculated by the corresponding PBGs based on the following formula:

$$D = \lambda_{max} / [1.63 \times (2.13 - \sin^2\theta)^{1/2}] \qquad (3)$$

For FCC structured PS@SiO_2 PCs, the PBG of could be theoretically calculated based on the following formuls[2]:

$$\lambda_{max} = 1.633D(n_{avg}^2 - \sin^2\theta)^{1/2} \qquad (1)$$

$$n_{avg}^2 = 0.74 \times n_{core\text{-}shnell}^2 + 0.26 \times n_{air}^2 \qquad (4)$$

$$n_{core\text{-}shell} = n_{core} X_{core} + n_{shell} X_{shell} \qquad (5)$$

Where λ_{max} is the maximum reflected wavelength, D represents the diameter of PS@SiO_2 core-shell microspheres, n_{avg} is the effective refractive index, θ is the incident angle; here n_{core} and n_{shell} are the refractive indexes of each component, $n_{core} = n_{PS}$ = 1.59, $n_{shell} = n_{silica}$ = 1.46, and n_{air} = 1.0; X_{core} and X_{shell} are the corresponding volume fractions in the PS@SiO_2 core-shell microspheres, respectively. The theoretical diameter of the prepared PS@SiO_2 core-shell microspheres can be calculated by the corresponding PBGs based on the above formulas.

Table S2. The diameter of the PS@SiO_2 microspheres (theoretical and practical)

	λ_{max} (nm)	θ (°)	Theoretical D (nm)	Practical D (nm)
PS	400	5	168	166
PS	441	5	185	187
PS	540	5	227	230
PS	552	5	232	233
PS@SiO_2	471	5	204	205
PS@SiO_2	509	5	220	221
PS@SiO_2	605	5	260	259
PS@SiO_2	733	5	322	324

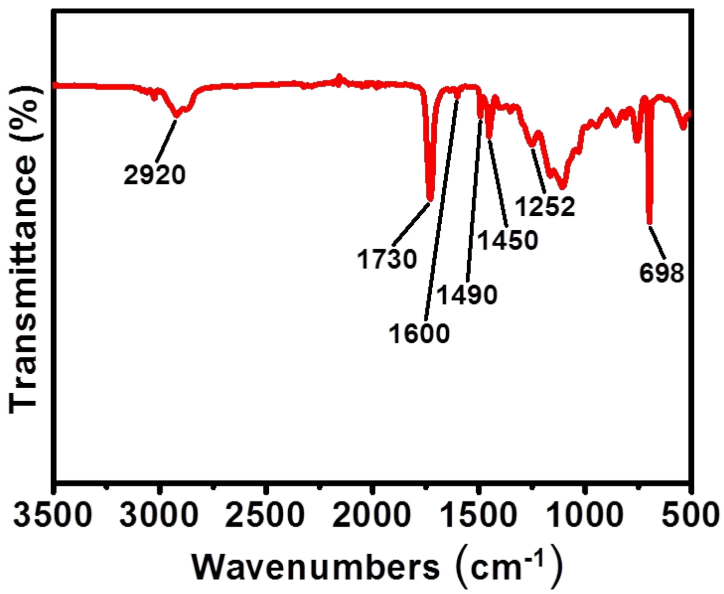

Figure S4. FT-IR spectra of the obtained DIOPC film.

PS: 2920 cm^{-1} (ν C-H aliphatic); 1600, 1490, and 1450 cm^{-1} (ν C-C framework of the benzene ring), 698 cm^{-1} (δ single replace of Benzene). Polymer backbone: 1730 cm^{-1} (ν C=O) and 1252 cm^{-1} (ν C-O-C).

Table S3. Physical parameters of the liquids used to verify the infiltration response of the patterned sample from nonwetted to wetted states.

Name	Molecular formula	Refractive index	Surface tension (mN/m)
Water	H_2O	1.33	72.8
Formamid	$HCONH_2$	1.45	58.2
Diiodomethane	CH_2I_2	1.74	50.8
Ethanol	C_2H_5OH	1.36	22.1

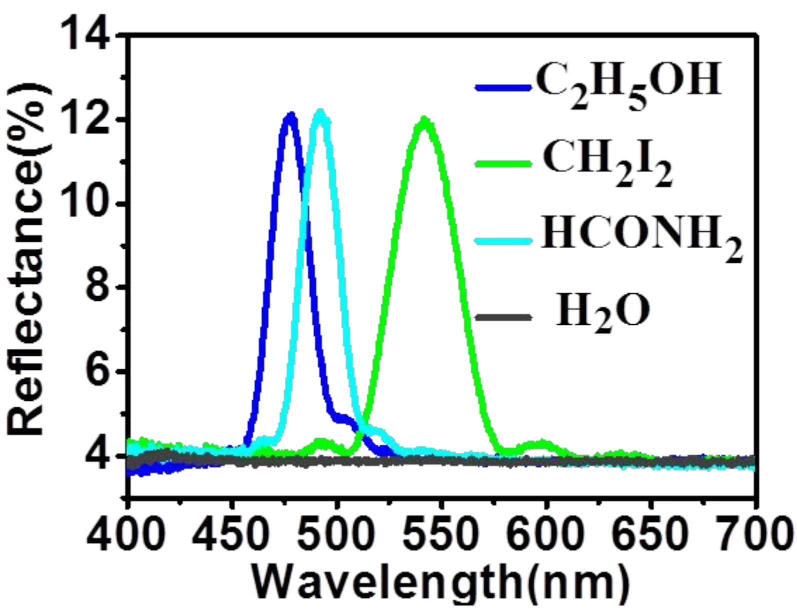

Figure S5. Corresponding reflection spectra of Figure 2d.

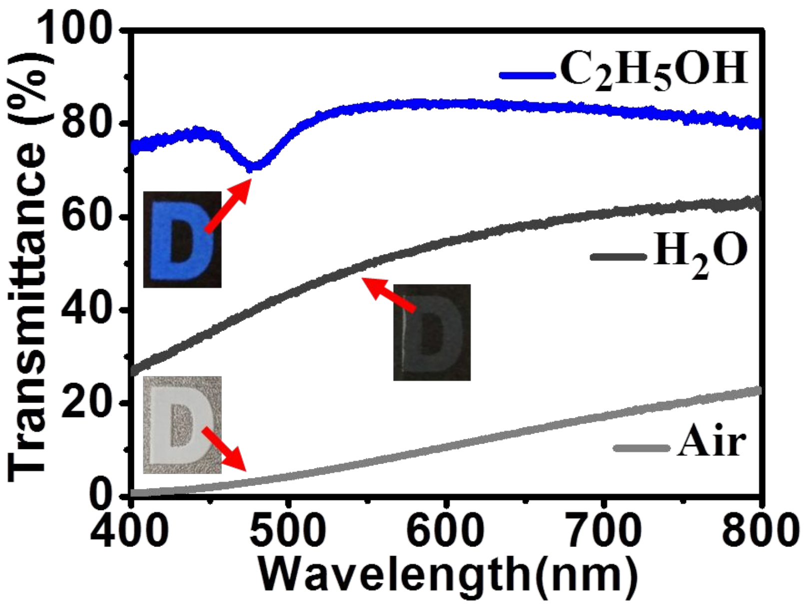

Figure S6. Transmission spectra of the prepared patterned DIOPC film with a letter "D" in the air or wiped with water or ethanol.

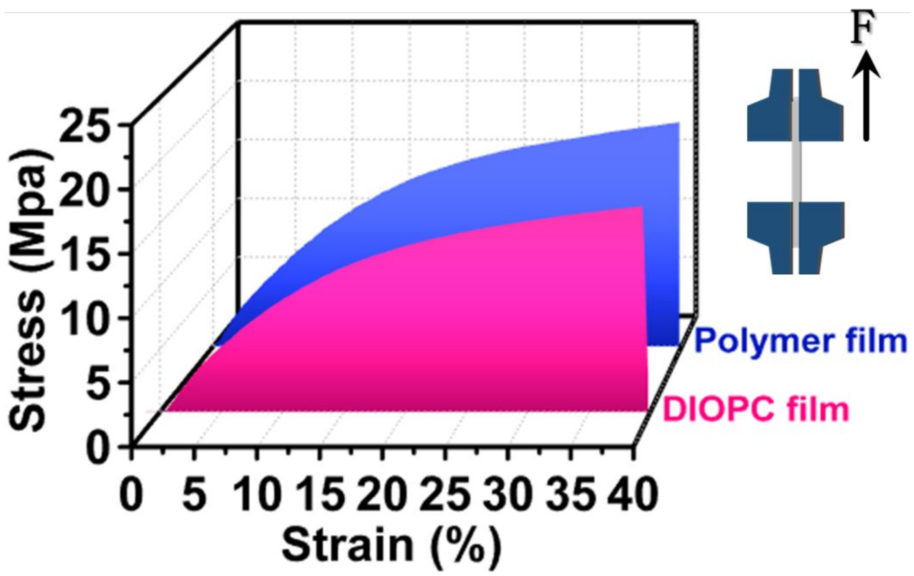

Figure S7. Tensile strain curves of the pure polymer film and the DIOPC film; the inset is the schematic diagram of the tensile test.

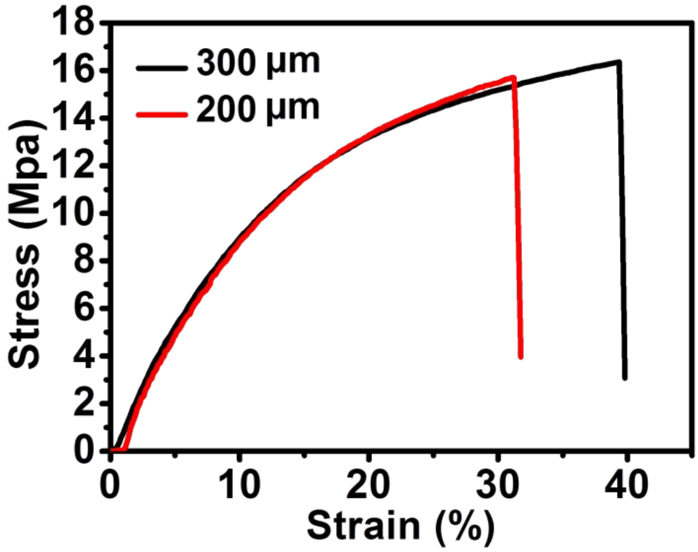

Figure S8. Tensile strain curves of the DIOPC film prepared with different thicknesses.

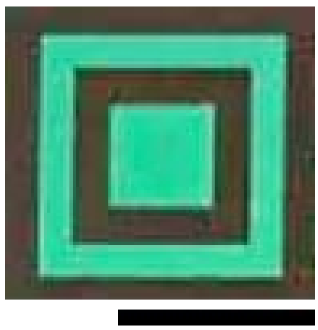

Figure S9. The pattern obtained by using the Fiber Laser Marking Machine. The scale bar in the inset is 0.5 cm.

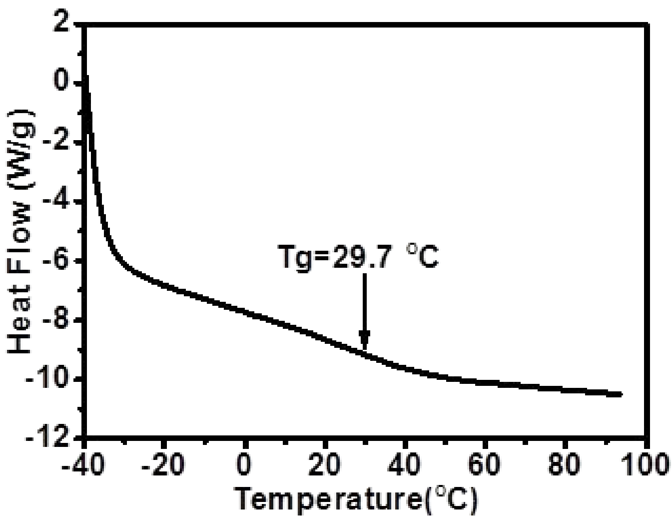

Figure S10. The DSC curve of the prepared BDIOPC film.

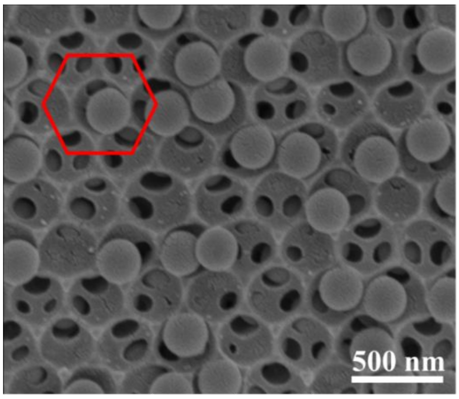

Figure S11. Cross-sectional SEM image of the close-up view of the top layer.

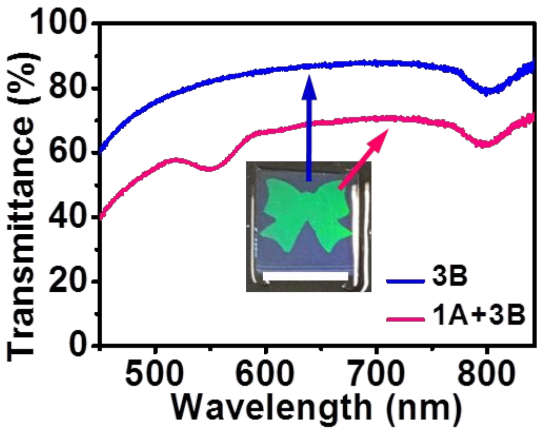

Figure S12. Transmission spectra of the prepared BDIOPC film sample wiped with ethanol both the top layer and bottom layer (3B represents the single top layer at 3B thickness and 1A+3B represents the whole BDIOPC film); inset is the photo of the BDIOPC film with a bowknot and the scale bar in the inset is 2 cm.

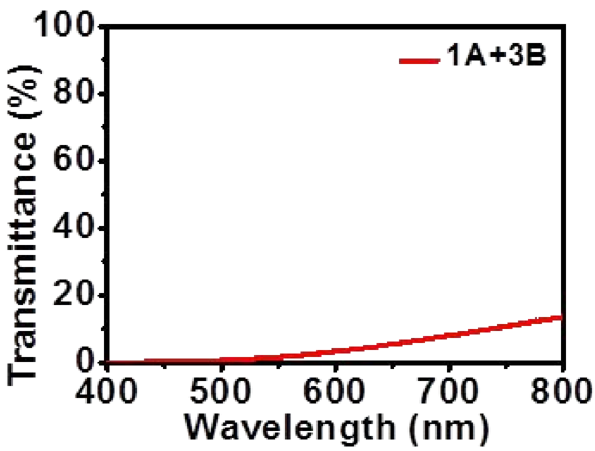

Figure S13. Transmission spectrum of the prepared BDIOPC film.

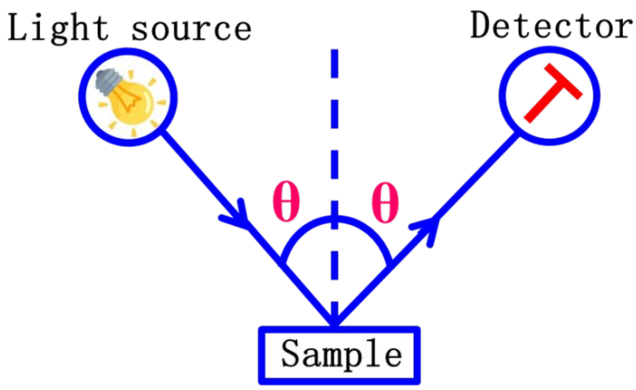

Figure S14. The diagram of the specular reflection model.

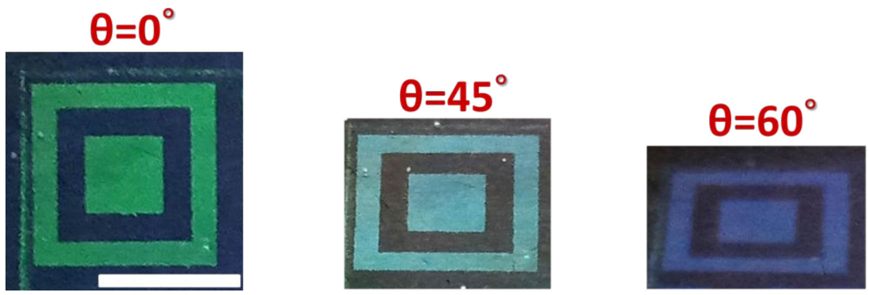

Figure S15. Digital photos of the patterned BDIOPC film at different viewing angles after the ethanol response. The scale bar in the inset is 0.5 cm.

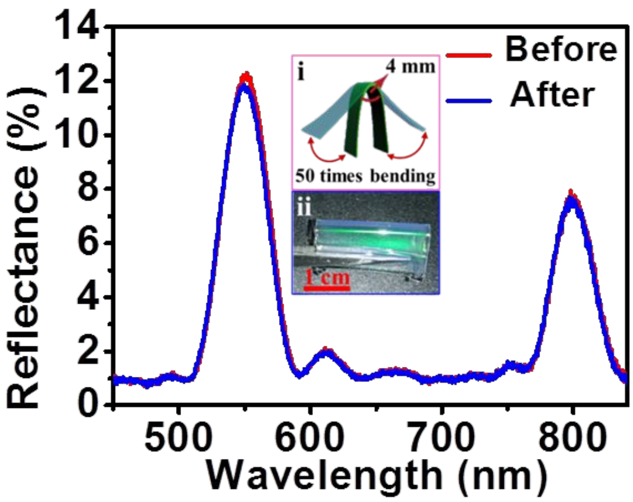

Figure S16. Reflectance spectra of the prepared BDIOPC film before and after 50 bending tests; insets are the schematic of the bending test and the obtained digital photo of the bent BDIOPC film.

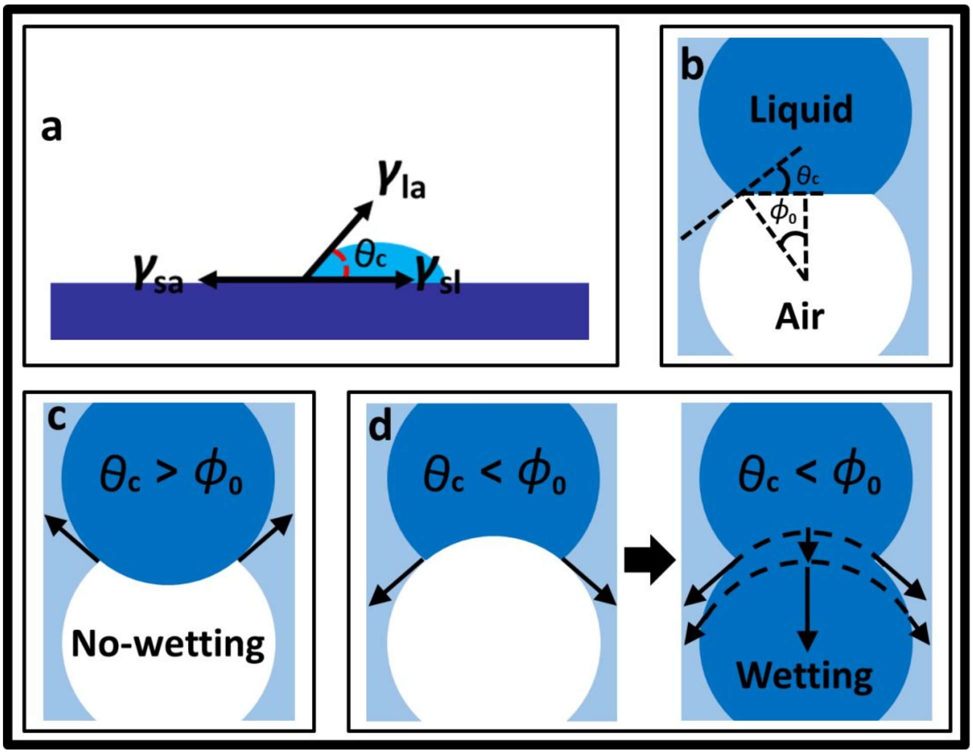

Figure S17. (a) Schematic diagram of the contact angle of the liquid on the solid surface ($\gamma_{sa} - \gamma_{sl} = \gamma_{la} \cos\theta_c$, where θ_c is the measured contact angle, γ_{sa}, γ_{sl}, and γ_{la} are the solid-air, solid-liquid, and liquid-air surface tensions, respectively). (b) Schematic of the liquid front propagating at the neck of two connected balls of the inverse opal structure (where θ_c indicates the measured contact angle of the liquid on the inverse opal structure surface, ϕ_0 is the azimuthal angle). (c) Schematic illustration of the no-wetting state (if $\theta_c > \phi_0$, no infiltration occurs). (d) Schematic illustration of the wetting state (if $\theta_c < \phi_0$, infiltration occurs).

	Top layer									
Ethanol concentrations (v/v%)	0	5	10	15	20	25	27.5	30	32.5	100
Contact angle	50.2°	42.8°	38.0°	34.1°	31.3°	26.9°	23.0°	18.6°	13.5°	1.5°

	Bottom layer									
Ethanol concentrations (v/v%)	0	5	10	15	20	22.5	25	27.5	30	100
Contact angle	47.0°	40.1°	36.2°	33.0°	28.9°	26.9°	25.5°	20.0°	16.0°	1.2°

Figure S18. Contact angles of different concentrations of ethanol on the surface of the obtained BDIOPC film.

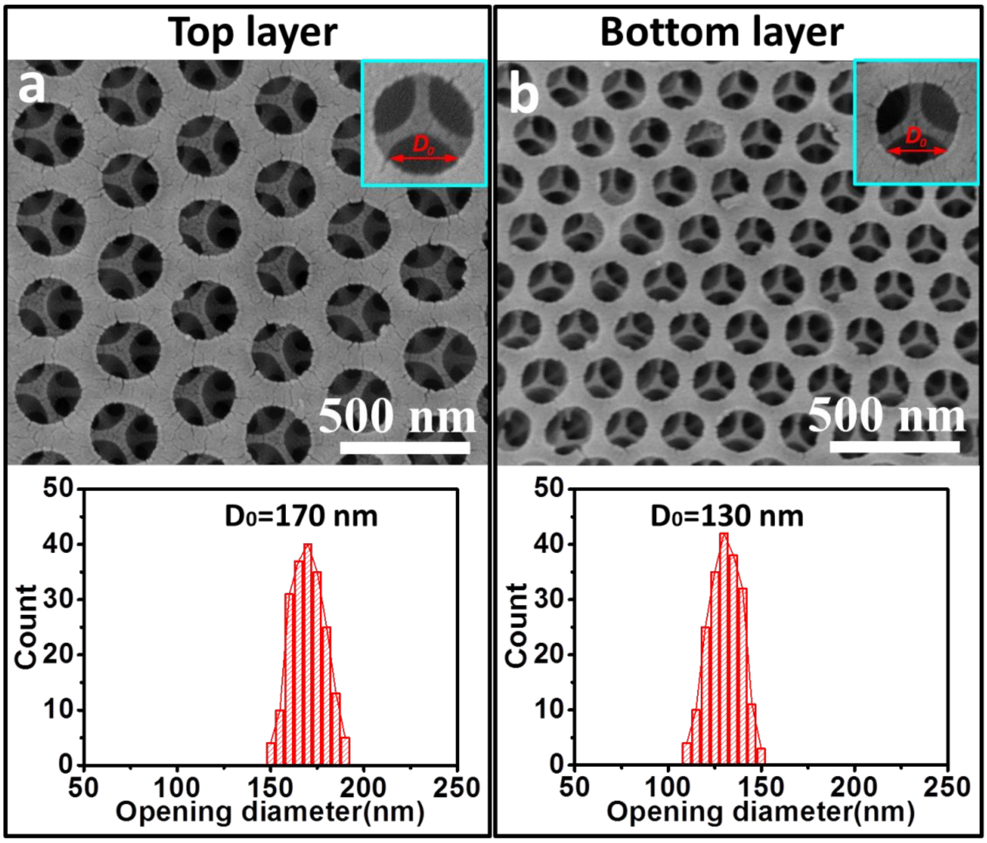

Figure S19. SEM images of the top view of the treated BDIOPC film with toluene (a, the top layer; b, the bottom layer; the PS spheres inside the pores of the BDIOPC film were removed) and corresponding opening diameter distribution diagrams.

References

(1) Aguirre, C. I.; Reguera, E.; Stein, A. Tunable Colors in Opals and Inverse Opal Photonic Crystals. *Adv. Funct. Mater.* **2010**, *20*, 2565-2578.

(2) Zhong, K.; Song, K.; Clays, K. Hollow Spheres: Crucial Building Blocks for Novel Nanostructures and Nanophotonics. *Nanophotonics*. **2018**, *7*, 693-713.