Supporting Information

Silica single-layer inverse opals: large-area crack-free fabrication and the regulation of transmittance in visible region

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Figures

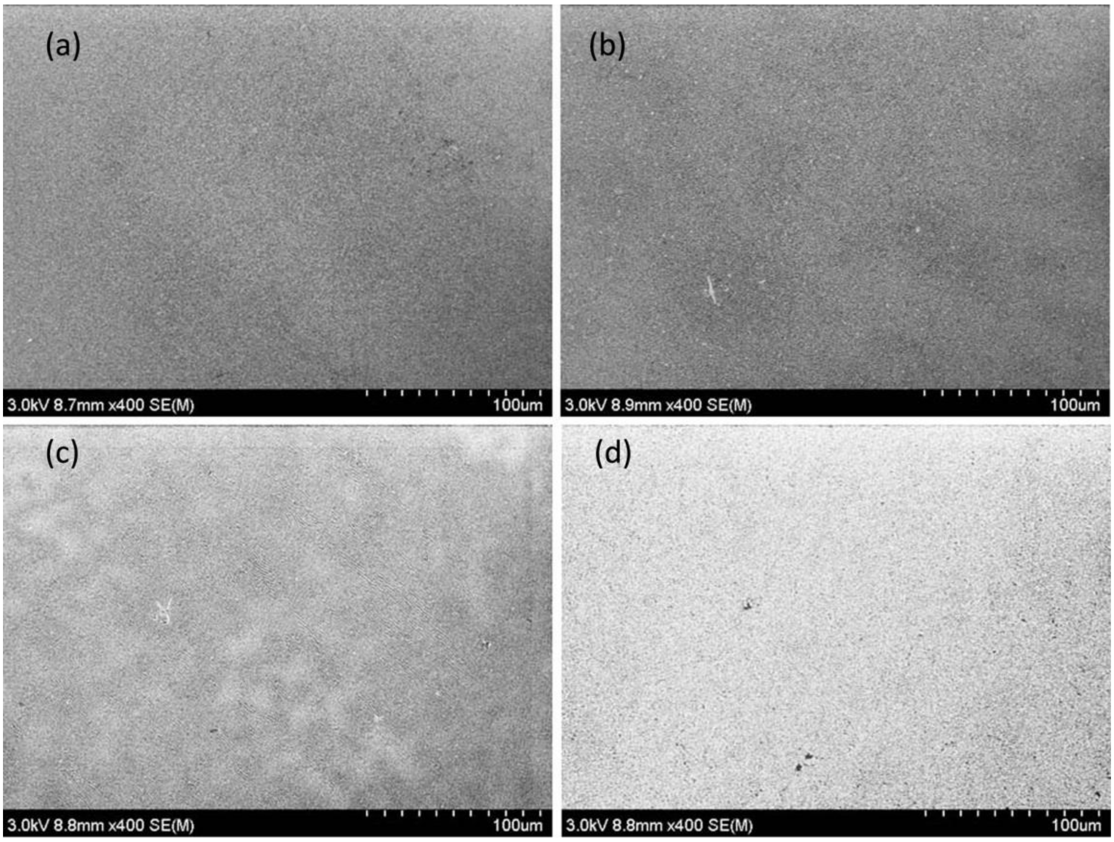

Figure S1 Low magnification SEM images of SiO_2 single layer IO films: (a) S220; (b) S410; (c) S530; (d) S930.

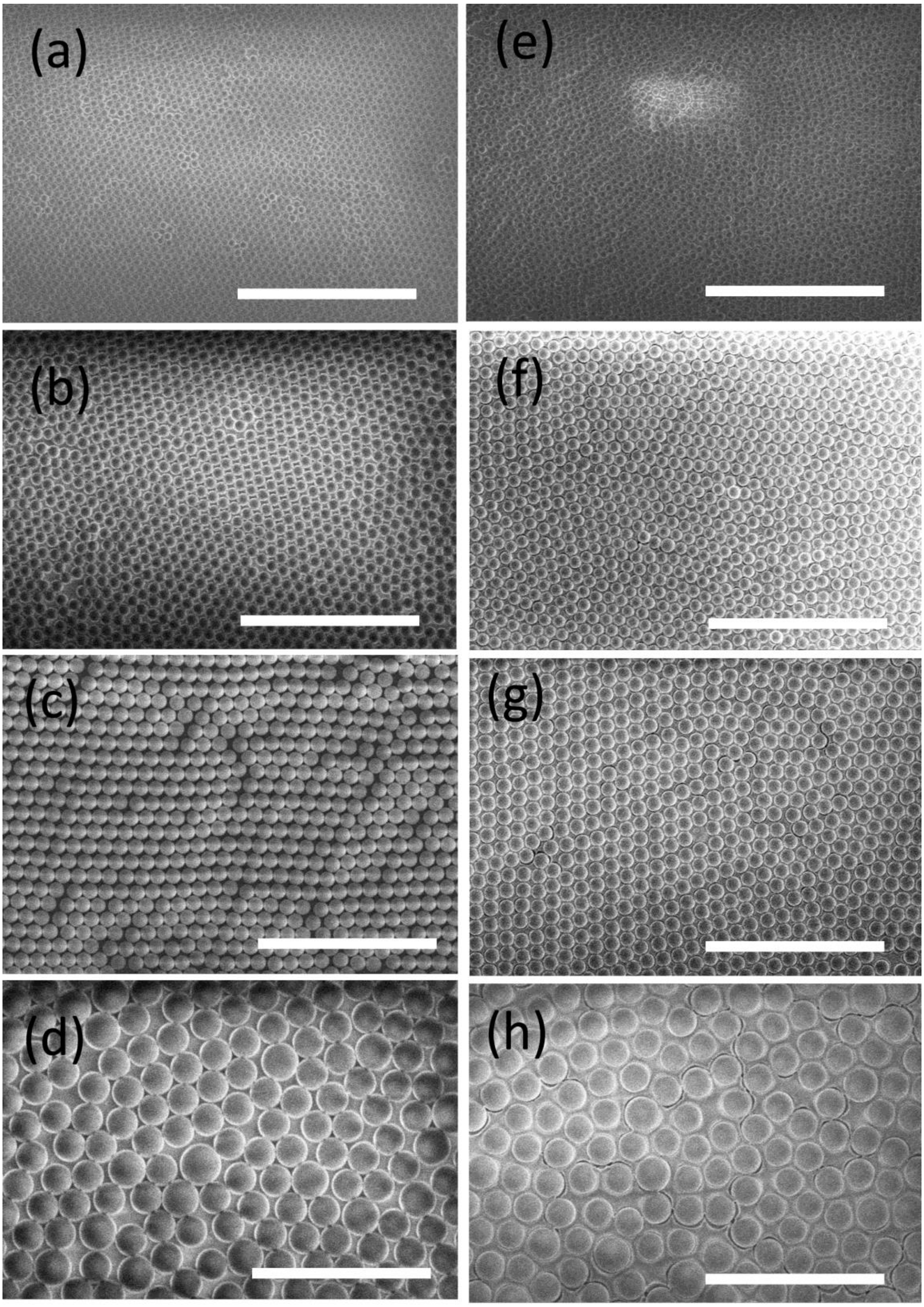

Figure S2 SEM images for the single-layer PS opal (a-d, left) and SiO_2/PS opal composite (e-h, right) obtained from 220 nm (a, e), 410 nm (b, f), 530 nm (c, g), 930 nm (d, h) PS spheres (scale bars are 5 μm).

Calculation S1:

Refractive index of PS opal & SiO_2/PS opal composite

Refractive index of PS opal series films

The prism-like unit structure of the PS opal films contains polystyrene and air and the effective refractive index of the film can be calculated using the following equation

$$n_{eff(PS\ opal+air)} = \sqrt{\frac{v_{PS}}{V_{Tot}} \times n_{PS}^2 + \frac{v_{air}}{V_{Tot}} \times n_{air}^2} \ , \qquad (S1)$$

where $\frac{v_{PS}}{V_{Tot}}$, $\frac{v_{air}}{V_{Tot}}$, n_{PS} and n_{air}, are respectively the volume ratios of polystyrene and air, and their corresponding refractive indexes. The total volume of the prism-like unit structure is given by:

$$V_{Tot} = area\ of\ trianglar\ base * height\ *\ D = \frac{1}{2} D^2 \sqrt{D^2 - (D/2)^2} \ , \qquad (S2)$$

where D is the PS spheres diameter as well as the opal period.
The volume of PS spheres of radius R contained inside a unit structure is simply half the volume of a sphere, or

$$3 \cdot \frac{1}{6} \cdot \frac{4}{3} \pi R^3 = \frac{2}{3} \pi R^3 \ . \qquad (S3)$$

Independently of the size of the PS spheres, the volume ratios of PS and air are the same for all PS series films and so are the refractive indexes of the films which are found to be 1.39.

Refractive index of SP series films

After the infiltration of SiO_2, the volume of the opal unit structure can be found using modified equation (S2). This time, because of infiltration, the period of the SiO_2/PS opal composite (see figure S3 bellow) is larger than the diameter of the PS spheres, and the volume of the unit structure is:

$$\frac{1}{2} D \sqrt{D^2 - (\frac{D}{2})^2} * \emptyset, \qquad (S4)$$

Where $\emptyset$ is the PS sphere diameter. Here the unit structure contains polystyrene, air and SiO_2, and the refractive index of the SP series can be calculated using the following equation:

$$n_{eff(PS+SiO_2+air)} = \sqrt{\frac{v_{PS}}{V_{Total}} \cdot n_{PS}^2 + \frac{v_{SiO2}}{V_{Total}} \cdot n_{SiO_2}^2 + \frac{v_{air}}{V_{Total}} \cdot n_{air}^2}, \qquad (S5)$$

where $\frac{v_{PS}}{V_{Total}}$, $\frac{v_{SiO2}}{V_{Total}}$, and $\frac{v_{air}}{V_{Total}}$, are respectively the volume ratios of the PS spheres,

SiO_2, and air and n^2_{PS}, n^2_{SiO2}, and n^2_{air} are their corresponding refractive indexes.
To find the volume of SiO_2 and air in the unit structure, and hence their volume ratios, we refer to Figure S1. At a certain height x, we consider a slice of thickness dx of the spheres taken through a plane normal to the figure. For a single sphere, this slice has a radius $r(x)$, so $r(x)^2 = R^2 - (R - x)^2$, and an area $\pi r(x)^2$. The volume of this slice is $dV = \pi r(x)^2 dx$. If the thickness of SiO_2 layer is "H", the volume of PS spheres included inside a portion of height H of the structural unit (half a sphere) is thus given by

$$V_{PS\ portion}(H) = ½ \int_0^H \pi r(x)^2 dx = ½ \int_0^H (R^2 - (R - x)^2)\ dx = ½\pi\,(RH^2 - H^3/3). \tag{S6}$$

Accordingly, the volume of SiO_2 is given by subtracting the volume of PS spheres portion from the prism portion of height H, that is,

$$V_{SiO_2} = V_{prism\ portion} - V_{PS\ portion}. \tag{S7}$$

The air and SiO_2 in the SP series opals occupy the space of the unit structure that has the looks of a golden cup. The volume of this golden cup is simply the volume of the PS portion in the prism-like unit structure subtracted from the total volume of the unit structure. The volume of air in the golden cup is thus

$$V_{air} = V_{goldcup} - V_{SiO_2}. \tag{S8}$$

The volume ratios can then be obtained by dividing the values from equations (S6), (S7), and (S8) by the total volume of the unit structure.

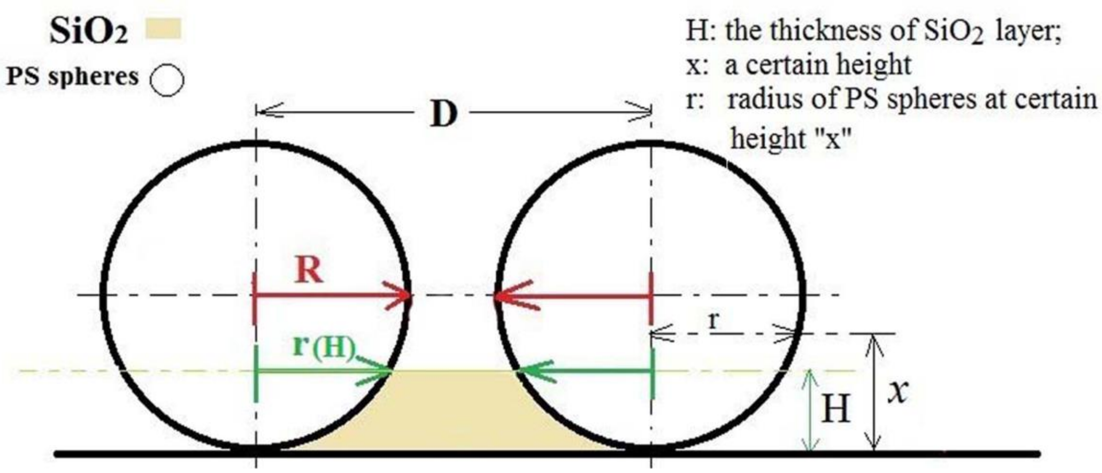

Figure S3 Illustration of SiO_2/PS opal composite.

Calculation S2

The Reflectance at the interface between film and substrate

Basically, when the incident light vertically travels from air to film, the interface reflectance could be calculated using the following equation derived from Fresnel's equations [see Ref.50 in the article]:

$$\mathbf{Ri_{SP}} = \left(\frac{\mathbf{n_1-n_2}}{\mathbf{n_1+n_2}}\right)^{\mathbf{2}} \tag{S9}$$

n_1: refractive index of one phase;
n_2: refractive index of other phase.

Calculation of the interface reflection between film and substrate of of SP and PS series:

Here, we consider two conditions. First, for PS opal, the incident light travels directly from pore to substrate, so the "n" should be that of air (=1.0); second, in the case of SiO_2/PS of the opal composite film, light travels from SiO_2 to substrate, so the "n" index should be that of SiO_2 (=1.42).

a) For SP series (i.e. SiO_2/PS opal composite film on substrate):
Here the interface between SiO_2 and the glass substrate, except for dot-contact between PS spheres and substrate, the interface consist mainly from the SiO_2 contacting with the substrate (n_1 = 1.51). So, the Ri_{SP} is determined by SiO_2 (n_0=1.42) and the substrate, calculated as follows:

$$Ri_{SP} = \left(\frac{n_1-n_0}{n_1+n_0}\right)^2 \times 100\ \% = \left(\frac{1.51-1.42}{1.51+1.42}\right)^2 \times 100\ \% = 9.4 \times 10^{-4}\ \% \tag{S10}$$

b) For PS series(i.e. PS opal film on substrate):
   Except for dot-contact between PS spheres and substrate, the interface mainly comes from air (from the pores) contacting with the substrate. So, the Ri_{PS} is, calculated as follows:

$$Rips = \left(\frac{n_1-n_0}{n_1+n_0}\right)^2 \times 100\ \% = \left(\frac{1.51-1.0}{1.51+1.0}\right)^2 \times 100\ \% = 4.1\ \%. \tag{S11}$$