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Electronic Supplementary Information

Dual-Colour Generation from Layered Colloidal Photonic Crystals Harnessing "Core Hatching" in Double Emulsions

Jun Hyuk Lee,[a] Gwan H. Choi,[a] Kyung Jin Park,[b] Dongjae Kim,[a] Juhyun Park,[c] Seungwoo Lee,[b] Hyunmin Yi,[d] and Pil J. Yoo*[ae]

[a] School of Chemical Engineering, Sungkyunkwan University (SKKU), Suwon 16419, Republic of Korea, *E-mail : pjyoo@skku.edu

[b] KU-KIST Graduate School of Converging Science and Technology, Korea University, Seoul 02841, Republic of Korea,

[c] School of Chemical Engineering and Materials Science, Chung-Ang University, Seoul 06974, Republic of Korea,

[d] Chemical and Biological Engineering Department, Tufts University, Medford, MA 02155, United States,

[e] SKKU Advanced Institute of Nanotechnology (SAINT), Sungkyunkwan University (SKKU), Suwon 16419, Republic of Korea.

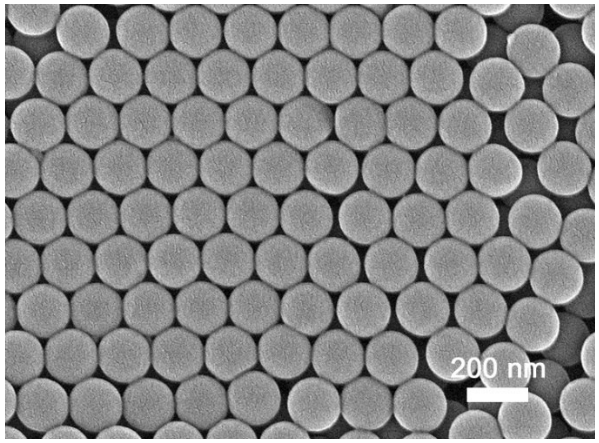

Figure S1. FE-SEM image of PVP-decorated PS nanoparticles. The size of PVP-decorated PS nanoparticles was analyzed to be 176.0±3.6 nm.

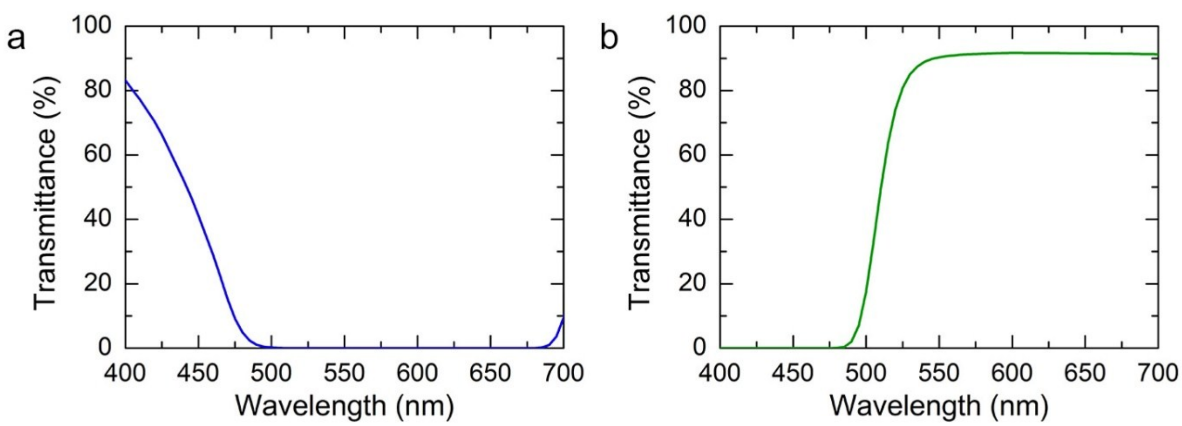

Figure S2. Light transmittance data of glass filters with (a) blue and (b) green (Thorlabs Coloured Glass Bandpass Filter FGB25 as blue colour filter and Coloured Glass Longpass Filter FGL515 as green colour filter).

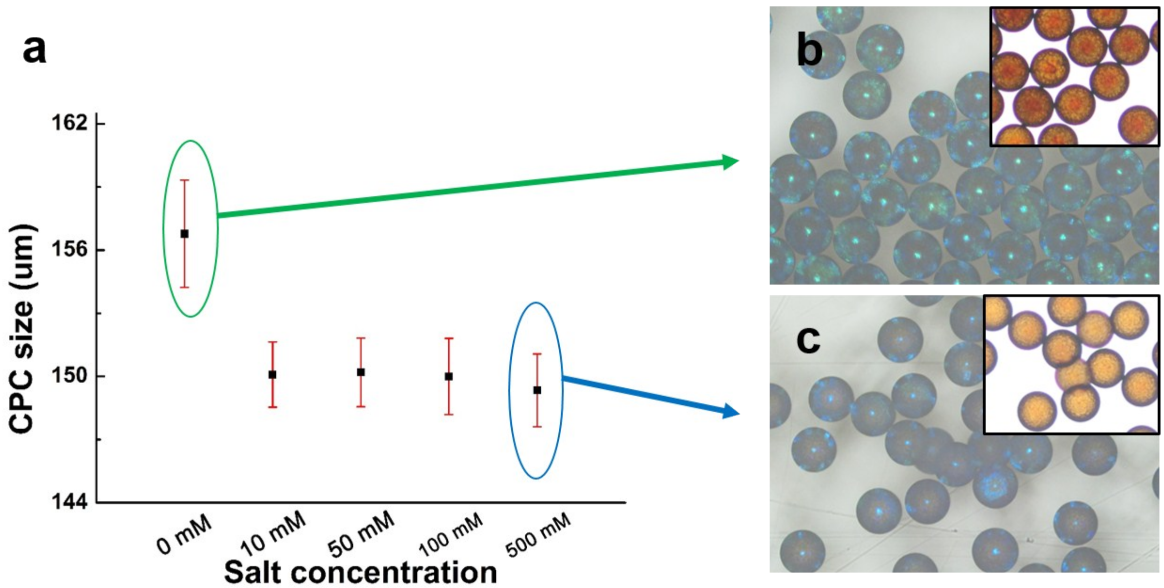

Figure S3. Size change of ethanol-swollen-CPC particles when re-dispersed in pure DI water and salt-dissolved water. (a) Sizes of CPC particles re-dispersed in salt-dissolved solution are smaller than that in DI water. (b) Dual-colours are observed from core-shell-like structured CPCs re-dispersed in pure DI water. (c) Due to intensified diffusion-out of ethanol solvent from CPCs in salt-dissolved water, CPC particles are strongly compressed and compacted to exert mono-colour generating characteristics.

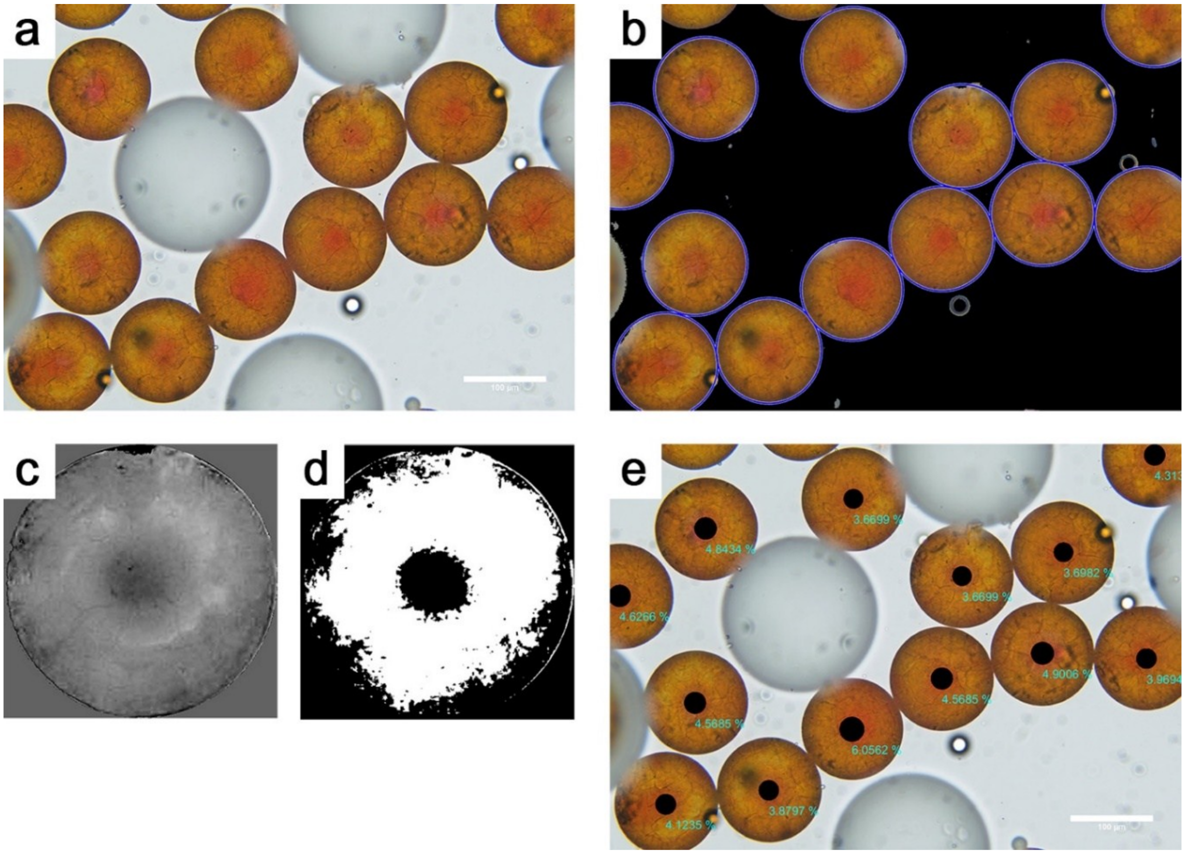

Figure S4. Procedures for estimating the areal ratio of red-centered core to the entire area of a single CPC particle. (a) First, an image of CPC particles was obtained in transmission mode, (b) The occupied regions with CPC particles were extracted from background using thresholding method, where pixels whose blue channel intensities were smaller than a manually chosen threshold were selected. In this example, the threshold value was 130 for 24-bit-coloured images. Then, size-invariant circle detection method[1] was used for separating each domain of CPC particles, which rendered blue circles of individual CPC particles, (c) Colour space transformation from RGB to HSV (hue, saturation, and value) was conducted to facilitate the segmentation of the centered core. (d) Otsu's method[2] was performed in the hue channel to identify the core region (coloured in black). Note that Otsu's method automatically found the optimum threshold value based on hue channel histogram. (e) Finally, red-centered core regions were identified and the areal ratio could be calculated for each CPC particle.

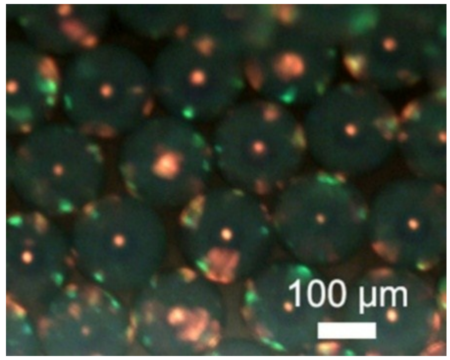

Figure S5. Core-shell-like CPC particles exhibiting dual colours of red (core) and green (shell) using 220 nm-sized PS nanoparticles.

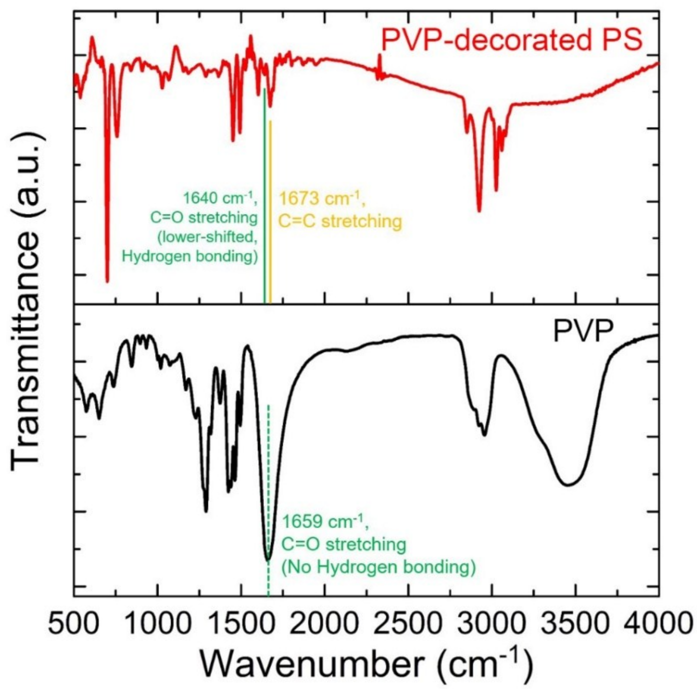

Figure S6. FT-IR spectral analysis of pure PVP (black) and PVP-decorated PS particles (red). The sample of PVP-decorated PS was prepared to be slightly wet to facilitate the formation of hydrogen bonding. While both PVP-decorated PS and PVP show C=O absorption peak, PVP-decorated PS exhibits lower-shifted peak from 1659 cm^{-1} to 1640 cm^{-1} due to the hydrogen bonding formation with protic solvent. Note that the peak emerged at 1673 cm^{-1} from PVP-decorated PS is originated from C=C stretching peak of aromatic ring of styrene.

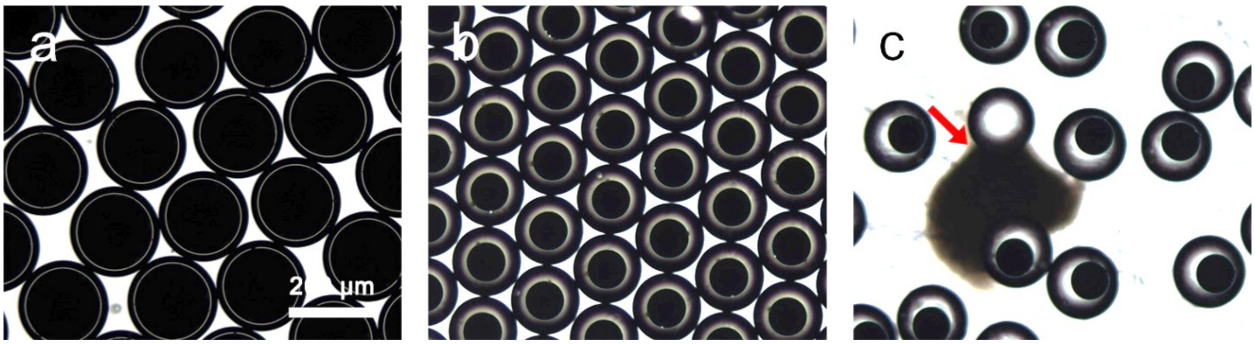

Figure S7. CPC generation using bare (non-PVP-decorated) PS nanoparticles. (a) Formation of double emulsions. (b) Crystallized CPCs after osmotic annealing process. (c) When internally stored CPCs were hatched by interfacial instability, however, the crystallized structure was burst and PS nanoparticles were completely dispersed into the water due to the absence of interlinking agents between particles.

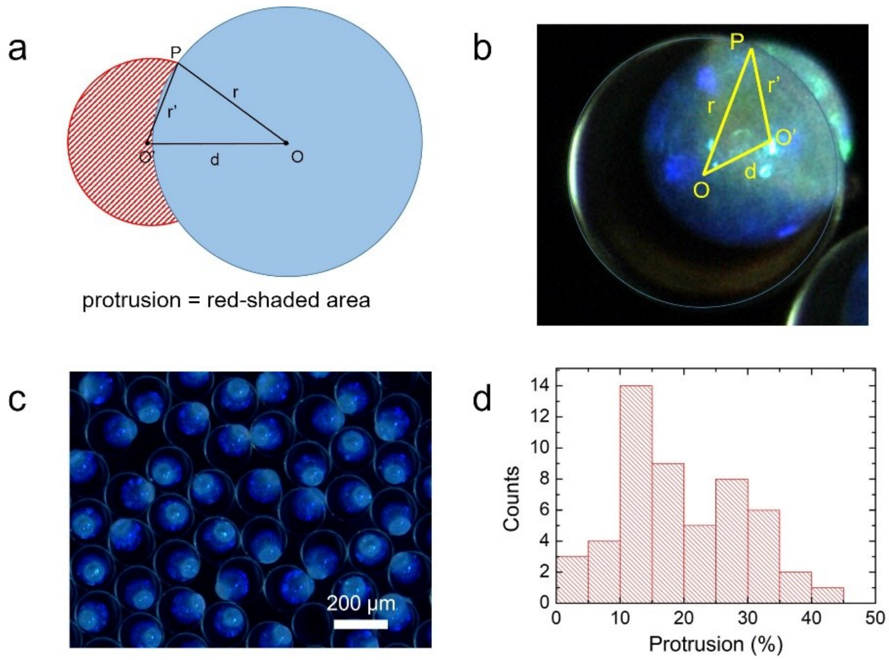

Figure S8. Geometric analysis of Janus-like CPC particles. (a) Schematic illustration of Janus-like CPC particle defined by three-lengthwise parameters of *r, r'*, and *d*. (b) Defining *r, r'*, and *d* from an optical microscopic image of Janus-like CPC. (c) Low magnification optical microscopic image of collected Janus-like CPCs. (d) Number distribution for an image of (c) showing the volumetric fraction of protruded region of CPC out of the cured ETPTA shell.

To quantify the structural uniformity of Janus-like CPCs, we have carried out the geometric analysis for the captured 47 particles, from which structural parameters of *r*, *r'* and *d* were obtained for estimating the volumetric fraction of protruded part in the CPC particles (Fig. S8). The volumetric fraction of the protruded region can be estimated by using following relationship,[3]

$$Volumetric\ protrusion = 1 - \frac{\frac{\pi}{12d}(r + r' - d)^2(d^2 + 2dr' - 3r'^2 + 2dr + 6rr' - 3r^2)}{\frac{4}{3}\pi r^3}$$

As statistically summarized in Fig. S8(d), there shows a distribution in the volumetric fraction of the partial protrusion, which is mostly placed in between 10~30%. Although the microfluidic system generates highly uniform double emulsions by means of elaborately controlling the flow rates, more or less irregularly localized UV-curing and subsequent particle hatching process would rather result in a marginal non-uniformity in the shape of Janus-like CPC particles.

REFERENCES

[1] Atherton, T.J.; Kerbyson, D.J. *Image and Vision computing* **1999**, 17**,** 795-803.

[2] Otsu, N. *Automatica*, **1975**, 11, 23-27.

[3] E. W. Weisstein, Sphere-Sphere Intersection, from a wolfram web source.