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Supporting Information

Light Diffusing, Down-converting Perovskite-on-Polymer Microspheres

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Experimental Section

Materials Preparation: Commercial highly crosslinked light-diffusing polymer (polystyrene, PS or polymethylmethacrylate, PMMA) microspheres with particle sizes ranging from 1 to 8 μm were received as free samples from Sekisui Plastics, Techpolymers. CsCl (99.999%), $PbCl_2$ (99.999%), CsBr (99.999%), $PbBr_2$ (99.999%), CsI (99.999%), DMF (extra dry, 99%) and Dimethyl sulfoxide (DMSO) were all purchased from Sigma-Aldrich, while PbI_2 (99%) was purchased from ACROS Organics^{TM}. Unless stated otherwise, all materials were used as received.

Preparation of Perovskite-polymer Microspheres: Perovskite precursors, CsX and PbX_2 (X= Br^-, I^-) with a molar ratio of 1:1 and an overall concentration of 0.04 mmol/ml were prepared in DMF solvent and stirred to be fully dissolved, while CsCl and $PbCl_2$ was prepared in DMSO solvent, Oleylamine (OAm) and oleic acid (OA) were added into the perovskite precursor solution with a concentration of 10 μl/ml and 100 μl/ml, respectively, except that in $CsPbI_3$ preparation, only OAm was used. The commercial light diffusing polymer microspheres were mixed with the prepared precursor solution at a concentration of 500 mg/ml. Subsequently, antisolvent (e.g. isopropyl alcohol (IPA)) was quickly injected into the stirring dispersion in nitrogen filled glovebox within 5 seconds to induce the formation of PPMs. The suspension was then centrifuged (4000 rpm) for 5 min and the precipitate was collected and dispersed again in IPA solvent. After another centrifugation, the supernatant was discarded and the precipitate was vacuum-dried overnight to obtain final PPMs.

PDMS embedded PPMs Fabrication: SYLGARD® 184 Silicon Elastomer Kit was used in this PDMS matrix formation. PPMs were added into the silicon elastomer base with a weight ratio of 1:20 (PPMs: silicon elastomer base) in a glass container. After ultrasonication for one hour to fully disperse microspheres in the elastomer base, the curing agent was then dropped into the container with a volume ratio of 1:7 (curing agent: base set). After stirring for 5 minutes, the glass container was vacuum-dried overnight, and then baked at 80℃ for 4 hours so that a homogeneous PPMs-in-PDMS down converting sheet was finally formed.

Characterization: Powder X-ray diffraction (XRD) patterns were performed on a PANalytical Empyrean X-ray diffractometer at 45 kV and 40 mA using Cu Kα radiation ($\lambda = 1.5406\dot{A}$). Spectra of PPMs were measured by Ocean Optics Spectrometer USB 2000+. The fluorescence images were taken using an Olympus BX51 microscope, the light source of 450-480 nm was used for excitation. Scanning electron microscope characterizations were carried out with Zeiss Ultra-55 FEG SEM. All samples for SEM measurements were coated with Au (5-10 nm) by sputtering. Transmission electron microscope characterization analyses were carried out using JEOL TEM-1011. All the characterizations were carried out with equipment Nanoscience Technology Center (NSTC) and Materials Characterization Facility (MCF) AMPAC of University of Central Florida. PLQY of samples were measured in a FluoroMax Plus (Horiba Jovin Yvon) spectrofluorometer equipped with the Quanta-Phi integrating sphere.

Half value angle (HVA) test: The HVA measurement was conducted with a goniometer, the PPMs-in-PDMS film was fixed at the goniometer central axis. With 450-nm blue LED as the exciting backlight and LS-110 Luminance Meter as the detector, the goniometer measures the scattered light luminance at different angles.

PPMs water stability test: 10 mg of green and red luminescent PPMs were dropped into 3~5ml deionized water in a test vial and keep storage in dark room ambient condition. While keeping the UV light, test vial and Ocean Optics Spectrometer at a fixed relative position, PL intensity changes were recorded. The boiling water test video was taken by simply adding 10 mg of PPMs into 3~5ml boiling deionized water.

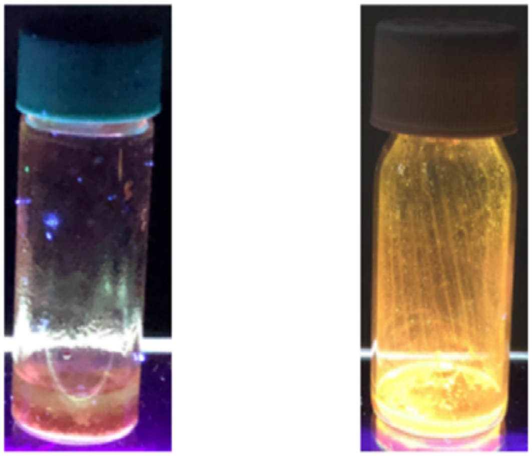


Figure S1. UV light illumination on $CsPbBrI_2$ nanocrystals prepared without PS microspheres (left) and with 8-μm PS microspheres (right).

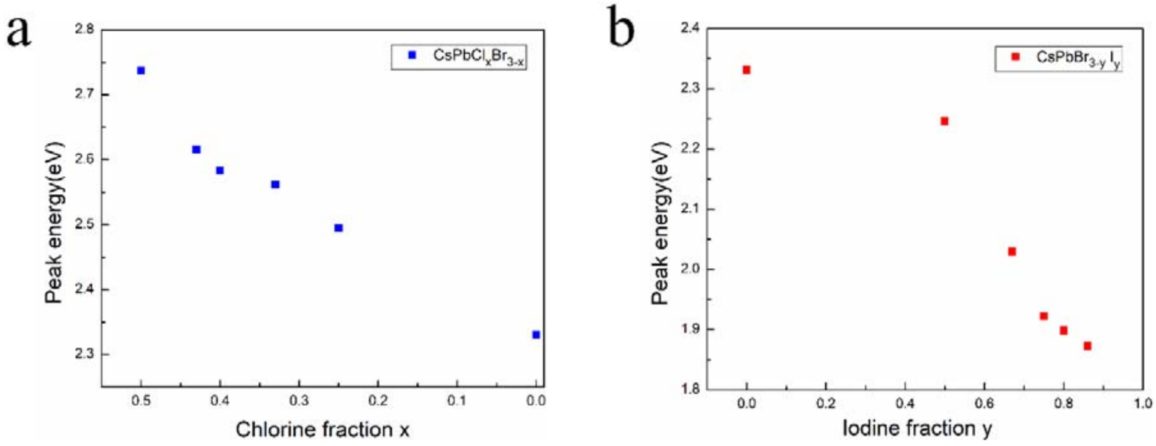


Figure S2. PL peak energies as a function of the anion composition fraction of a) Chlorine in $CsPbCl_xBr_{3-x}$ and b) Iodine in $CsPbBr_{3-y}I_y$.

Table S1. Emission peaks, FWHMs and corresponding CIE1931 color coordinates of selected PPMs.

PPM components	**Emission peaks (nm) (UV excitation)**	**FWHM (nm)**	**CIE color coordinates (x,y) (Blue LED excitation)**	**PLQY (%)**
$CsPbCl_{1.5}Br_{1.5}$	453	12.5	-	-
$CsPbCl_{1.2}Br_{1.8}$	480	18.7	(0.096, 0.260)	-
$CsPbCl_1Br_2$	484	18	(0.074, 0.532)	-
$CsPbBr_3$	532	22.6	(0.170, 0.763)	70.6
$CsPbBr_{1.5}I_{1.5}$	552	35.8	(0.416, 0.578)	-
$CsPbBr_1I_2$	611	32.7	(0.391, 0.600)	-
$CsPbBr_{0.75}I_{2.25}$	645	29.2	(0.662, 0.338)	40.5
$CsPbBr_{0.6}I_{2.4}$	653	36.5	(0.668, 0.332)	-
$CsPbBr_{0.42}I_{2.58}$	662	36.5	(0.719, 0.281)	-

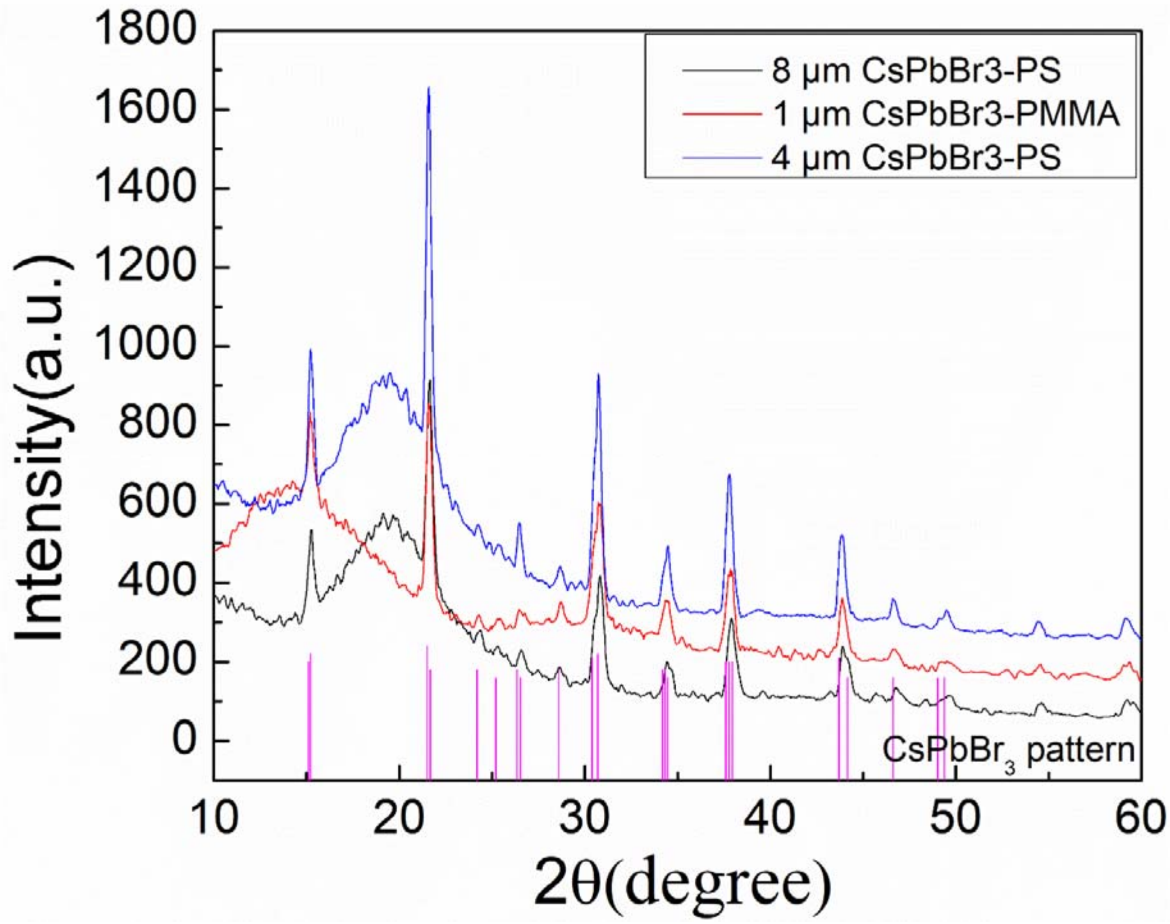


Figure S3. XRD patterns of different CsPbBr$_3$-polymer systems.

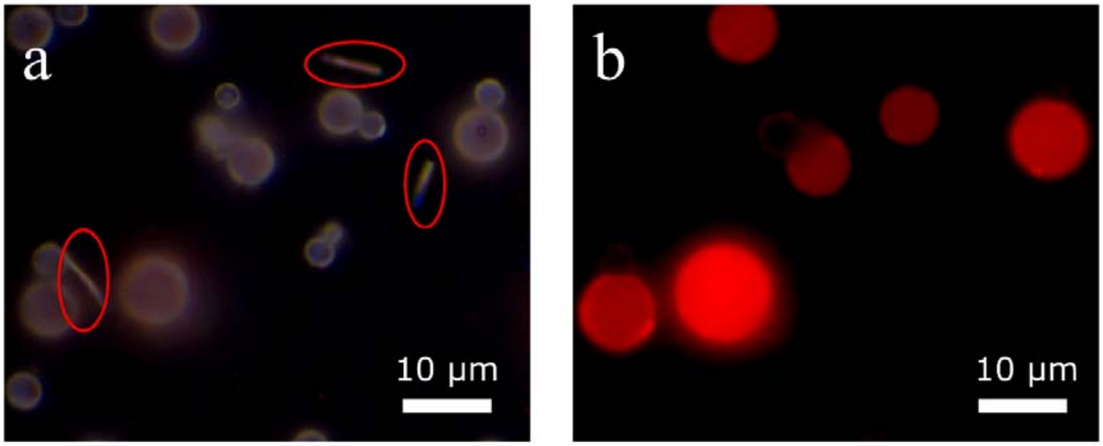
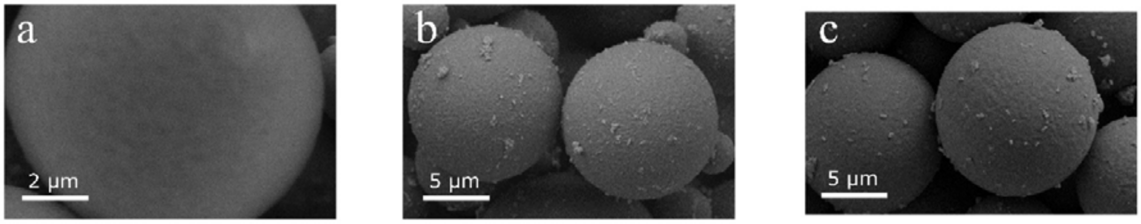


Figure S4. a) Bright field and b) Fluorescent optical microscope images of $CsPbBr_xI_{3-x}$-PS microspheres (x=0.75) with an average size of 8 μm, the rod shaped non-luminescent microcrystals was marked with circles.



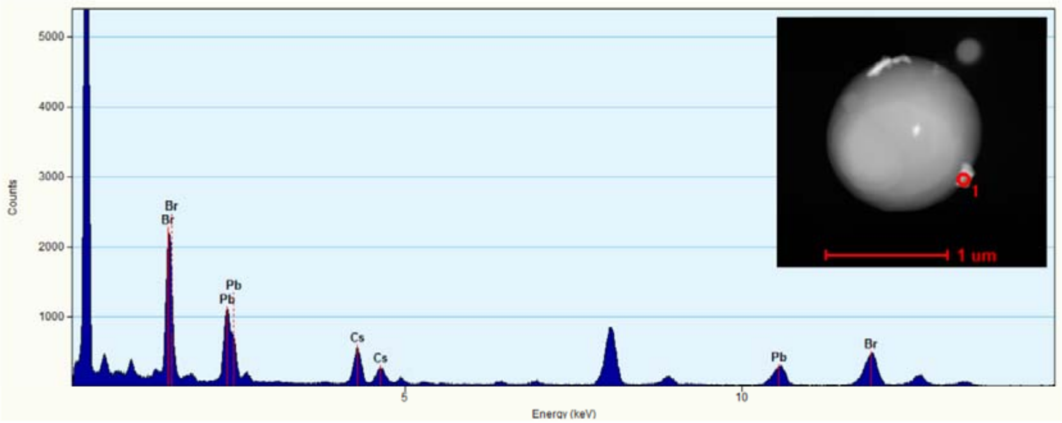
Blank PS microsphere

$CsPbCl_1Br_2$-PS microspheres

$CsPbBr_{0.75}I_{2.25}$-PS microspheres

Figure S5. SEM images of a) blank 8-µm PS microspheres , b) $CsPbCl_1Br_2$ –PS microspheres, and c) $CsPbBr_{0.75}I_{2.25}$ –PS microspheres.

a



b

Element	Weight(%)	Atomic(%)	Uncert.(%)	Detector Correction	k-Factor
Cs(L)	22.16	19.08	0.25	0.98	2.993
Pb(L)	34.73	19.18	0.37	0.99	4.409
Br(L)	43.09	61.72	0.40	0.92	2.638

Figure S6. EDS characterization of a single PPM. a) EDS spectra taken from highlighted region 1 ($CsPbBr_3$) for the upper image, respectively, showing the existence of element Cs, Pb and Br in the anchored nanocrystals. b) Table showing the Cs, Pb and Br atomic ration analysis from point 1.

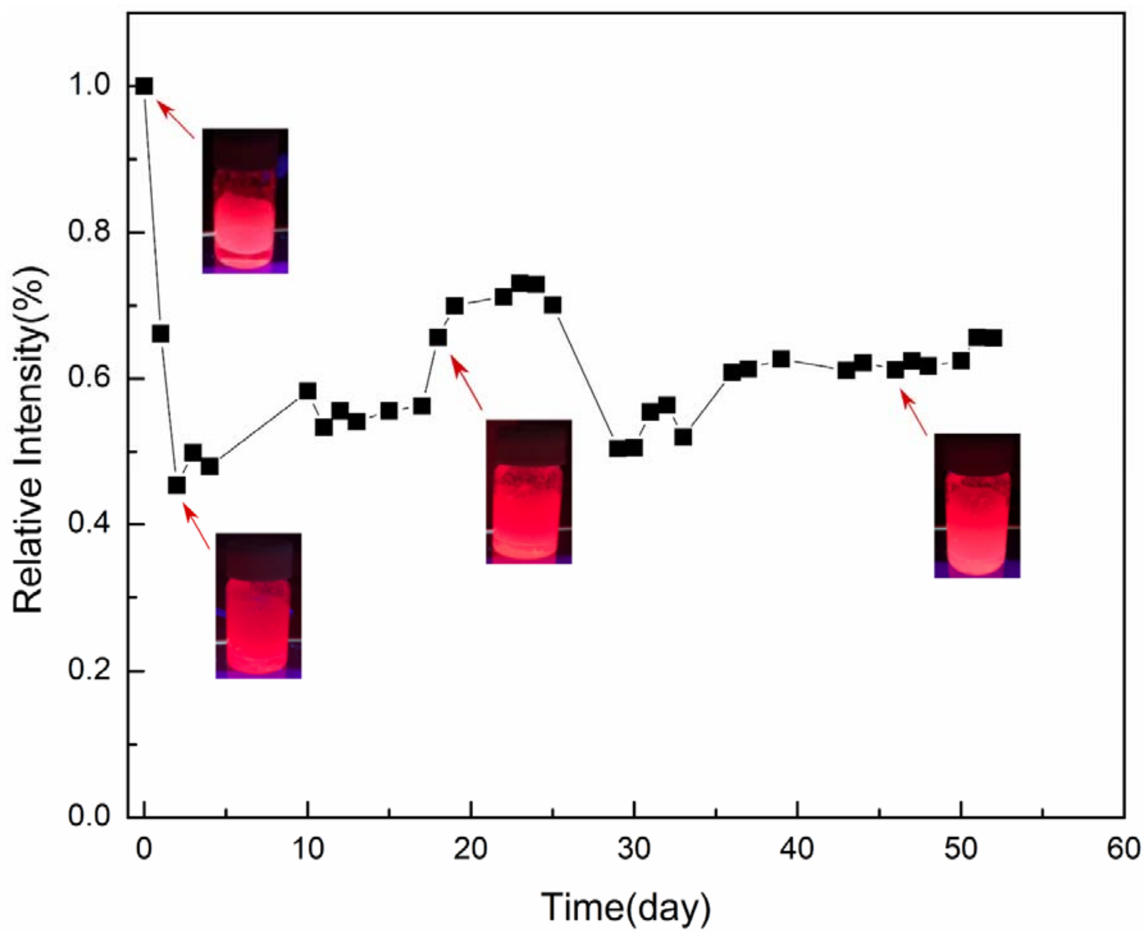


Figure S7. PL intensity changes of the $CsPbBr_{0.75}I_{2.25}$-PS microsphere immersed into water for over 50 days.

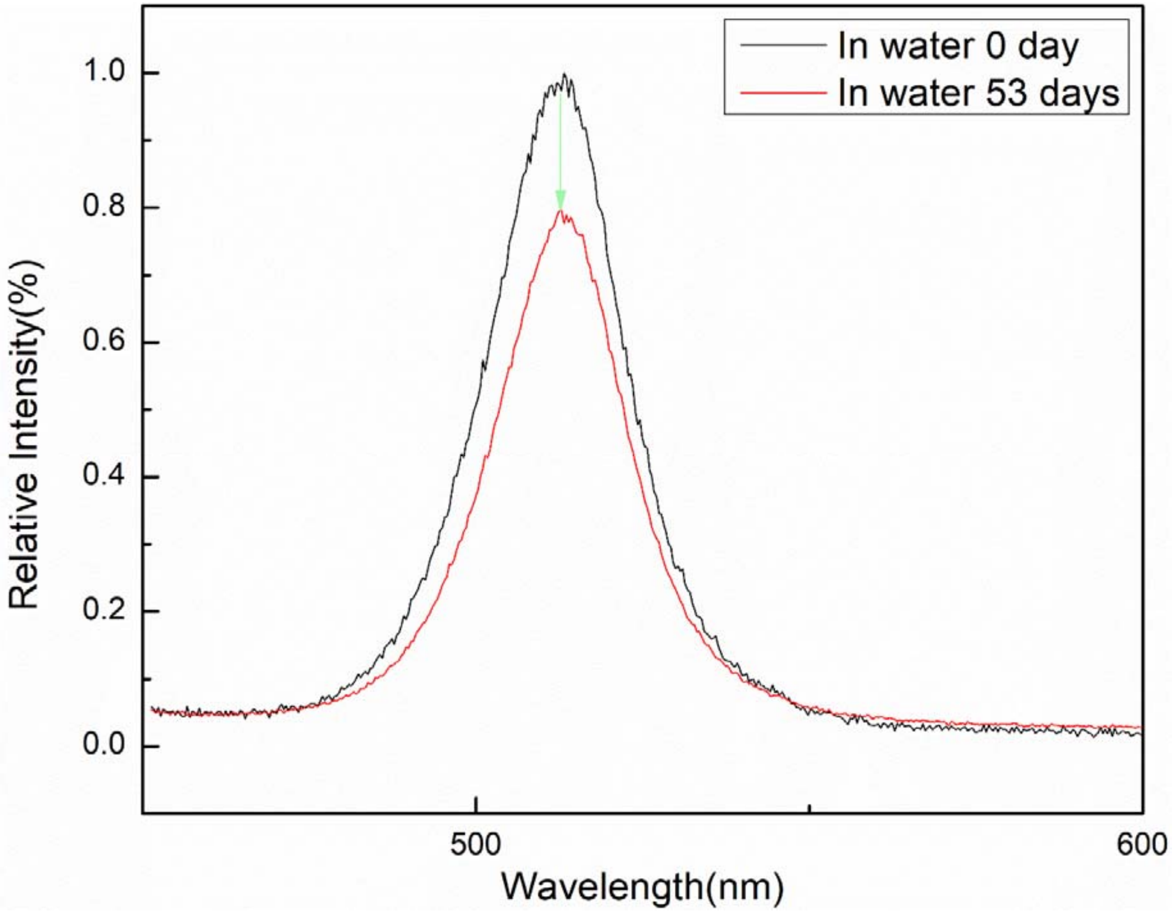


Figure S8. The relative PL intensity of the as-synthesized $CsPbBr_3$-PS microspheres before and after immersion into water for 53 days.

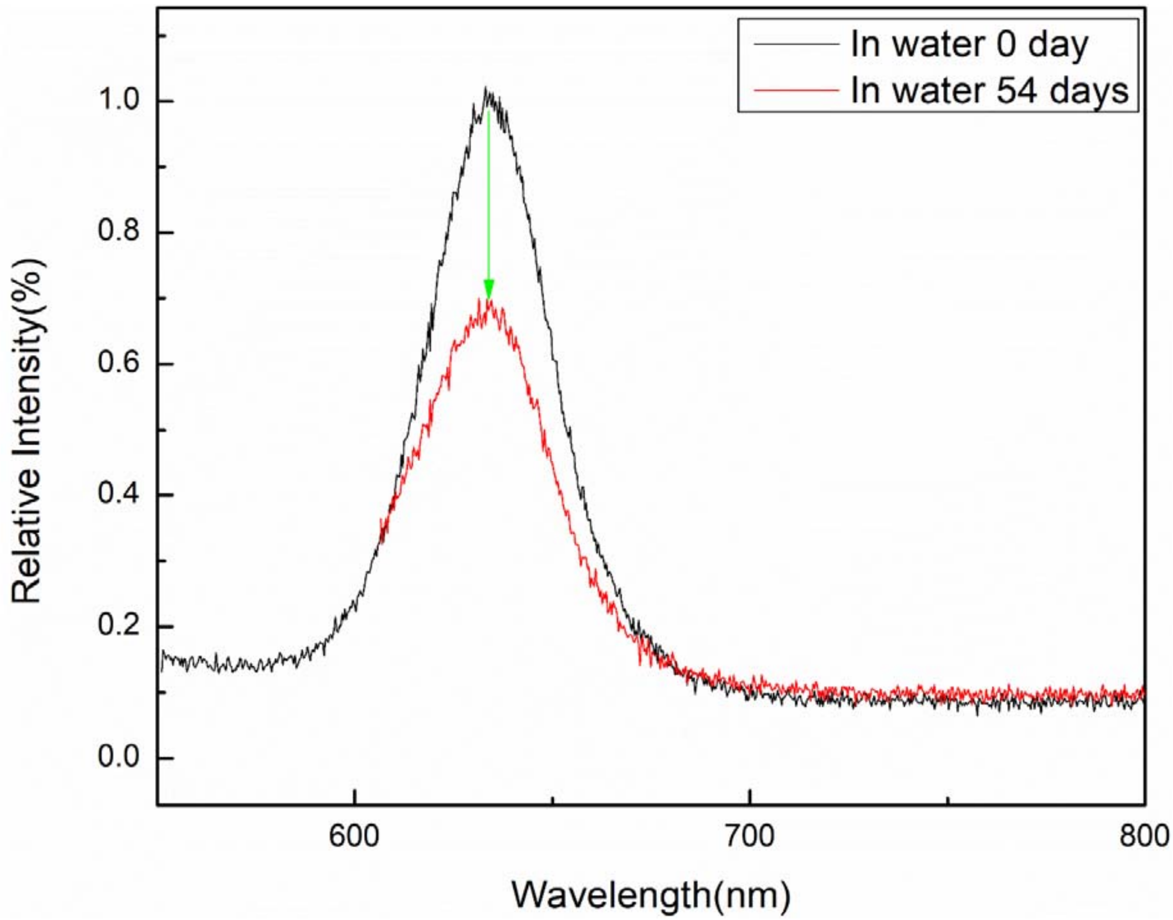


Figure S9. The relative PL intensity of the as-synthesized $CsPbBr_xI_{3-x}$-PS (x=0.75) microspheres before and after immersion into water for 54 days.

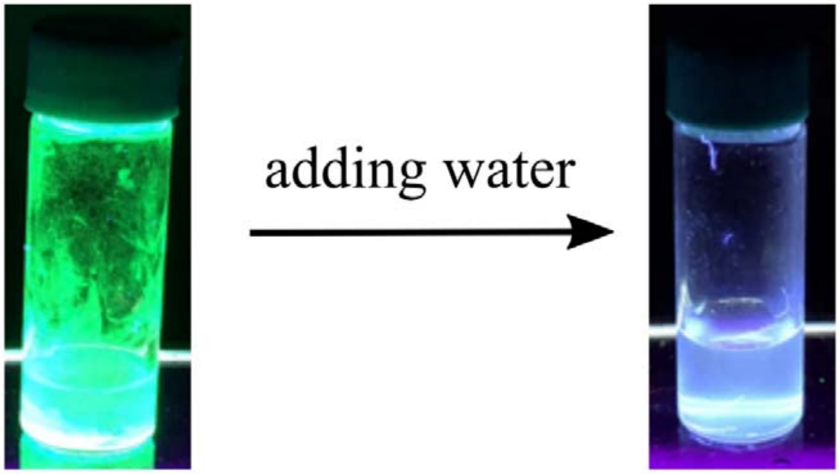


Figure S10. UV light illumination on $CsPbBr_3$ nanocrystals prepared without light-diffusing microspheres before (left) and after immersion in water (right).

Video S1. PPMs synthesis process.

Video S2. PPMs boiling water test.