

Supplementary Information for

Water-oil interfacial kinetics control enables water-based organic solar cells with 12.59% certified efficiency

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Supplementary Information Table of Contents

Supplementary Notes	3
Supplementary Figures	9
Supplementary Tables.....	57
Supplementary References.....	83

Supplementary Notes

Supplementary Note 1 | GSK classification of solvents

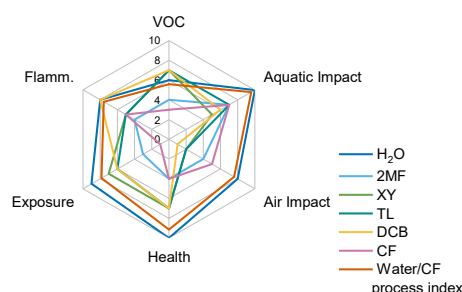
GlaxoSmithKline (GSK) solvent sustainability scores were used in this study to evaluate the environmental, safety and human-health implications of water and representative organic solvents commonly employed for active-layer fabrication. In the present analysis, category scores were normalized to a scale from 1 to 10, where 1 represents the least favorable performance and 10 the most favorable. Color coding followed the GSK solvent selection guide, with scores below 3 shown in red, scores between 3 and 7 in light yellow, and scores above 7 in dark green. The score of solvents was obtained directly from reference¹. We focus on six categories that are particularly relevant to printed active-layer manufacturing: volatile organic compound (VOC) emissions, aquatic impact, air impact, health hazard, exposure potential, and flammability & explosion. Together, these categories capture environmental, occupational and process-safety risks, and provide a practical basis for evaluating the need for additional engineering controls, such as VOC abatement, explosion-proof electrical systems and hazardous-waste segregation.

For overall solvent assessment, the solvents were further grouped into three classes. Hazardous solvents were defined as those showing either a severely unfavorable score (<2) or two red category scores. Problematic solvents were defined as those showing either one borderline score ($=3$) or two yellow category scores. The other solvents were classified as Recommended. This classification can provide an intuitive summary of the multidimensional GSK dataset while preserving the underlying category-specific information. Radar plots constructed from these six category scores highlight the comparatively low environmental and human-health burdens of water-based processing, and further suggest its potential to reduce solvent-specific industrial control requirements.

It should be noted that, although the final active layer is deposited from aqueous ink, the nanoparticle-synthesis step still involves chloroform. Therefore, to account quantitatively for chloroform consumption, we revised the solvent-sustainability assessment using the actual chloroform-to-water volume ratio of 1:6. For each assessment category, we calculated a volume-weighted GSK-derived index based on the relative processing volumes of the two liquids. The calculated values describe the relative impact of the solvents and do not represent a standard GSK score. Water and chloroform are therefore also reported separately to retain visibility of the intrinsic hazards associated with chloroform.

Solvent Name	Incineration	Recycling	Bioreatment	VOC Emissions	Aquatic Impact	Air Impact	Health Hazard	Exposure potential	Flammability & Explosion	Reactivity & stability	Life Cycle Analysis	Overall ranking
Water	4	2	4	6	10	8	10	9	8	10	10	Recommended
p-Xylene	10	9	6	7	5	2	7	7	5	10	7	Hazardous
Toluene	10	7	6	7	7	2	7	6	5	10	7	Hazardous
2-Methyltetrahydrofuran	6	5	3	4	7	4	4	3	4	6	4	Problematic
Tetrahydrofuran*	5	5	2	3	9	3	7	5	4	6	4	Hazardous
Chlorobenzene* ^a	4	9	7	7	7	2	4	4	8	10		Hazardous
Dichlorobenzene	4	8	6	7	6	1	7	6	8	10		Hazardous
Chloroform* ^a	3	9	5	3	7	5	4	1	5	10	6	Hazardous
CF/water process index ^b	3.9	3.0	4.1	5.6	9.6	7.6	9.1	8	7.6	10.0	9.4	Not classified

* Carcinogenic; ^a Reproductive harm. ¹ ^b Volume-weighted index of the water-chloroform mixture



Supplementary Fig. 1 | Radar plots of solvents GSK scores on VOC emissions, aquatic impact, air impact, health hazard, exposure potential, and flammability & explosion. 2MF denotes 2-Methyltetrahydrofuran. Flamm. denotes Flammability & Explosion. The GSK-derived process index was calculated for each category by weighting the chloroform and water scores according to their processing volume ratio of 1:6, which is used to describe the relative impact of the solvents and does not represent a standard GSK score.

Supplementary Note 2 | Determination of interfacial tension

The surface free energy of pristine PM6 and Y6 films was obtained from static contact-angle measurements using two probe liquids (water and diiodomethane) and analyzed using a standard two-liquid surface-energy approach. The surface tension of the aqueous phase (deionized water and 10 mg mL⁻¹ SDS solution) was measured by the pendant-drop method. Aqueous-organic interfacial tension was measured between chloroform-based organic solutions (with specified additive contents) and the SDS aqueous phase using the pendant-drop method and the drop profiles were fitted using the Young-Laplace equation.

The interfacial tension between a solid film (PM6 or Y6) and the SDS solution was estimated using Young's relation:

$$\gamma_{S-L} = \gamma_S - \gamma_L (\cos \theta_{S-L}) \quad (1)$$

where γ_S is the surface free energy of the PM6 or Y6 film, γ_L is the surface tension of the SDS aqueous solution, and θ_{S-L} is the contact angle of the SDS solution on the corresponding film.

The PM6-Y6 interfacial tension was calculated from the dispersive and polar components of the surface energy using:

$$\gamma_{PM6-Y6} = \left(\sqrt{\gamma_{PM6}^{disp}} - \sqrt{\gamma_{Y6}^{disp}} \right)^2 + \left(\sqrt{\gamma_{PM6}^{polar}} - \sqrt{\gamma_{Y6}^{polar}} \right)^2 \quad (2)$$

where γ_X^{disp} and γ_X^{polar} are the dispersive and polar components of the surface energy of material X, respectively.

Supplementary Note 3 | Characterization of residual SDS

For residual-SDS analysis, solids were collected from the unpurified dispersion and from the dispersion after five centrifugation-washing cycles and dried under identical conditions. FTIR spectra were recorded in KBr transmission mode. The C≡N stretching band of Y6 at approximately 2216 cm⁻¹ was used as an internal reference. Two SDS-associated bands near 592 and 1220 cm⁻¹ were analyzed independently. The former was treated as an empirical SDS fingerprint with relatively limited interference from PM6 and Y6, whereas the latter was attributed predominantly to sulfate-headgroup-related S-O vibrations.

The transmittance spectra were first converted to absorbance according to $A(\lambda) = -\log_{10}(T)$, where $A(\lambda)$ is the absorbance at a certain wavelength λ and T is the corresponding transmittance. Then, local linear baselines were constructed over the Y6 C≡N region and the two SDS-associated regions for a calculation of integrated areas of the bands near 2216, 592 and 1220 cm⁻¹ (denoted as S_{2216} , S_{592} , and S_{1220} , respectively). For each SDS-associated band, a normalized response was calculated using

$$R_{\text{SDS}} = \frac{S_{\text{SDS}}}{S_{2216}} \quad (3)$$

Assuming that the integrated absorbance ratio is proportional to the SDS-to-Y6 mass ratio and the mass loss of Y6 before and after centrifugation is negligible, an equivalent residual SDS concentration referenced to the initial aqueous-phase volume was estimated as

$$C_{\text{SDS},5} = 10 \frac{R_{\text{SDS},5}}{R_{\text{SDS},0}} \text{ mg mL}^{-1} \quad (4)$$

For the unwashed and 5-times-washing samples, the S_{2216} values were 0.24 and 0.78, respectively. The corresponding S_{592} values were 4.33 and 0.40, giving normalized responses of 18.04 and 0.513. The residual fraction estimated from the 592 cm⁻¹ band was therefore 2.84%, corresponding to an equivalent SDS concentration of about 0.28 mg mL⁻¹. For the band near 1220 cm⁻¹, the integrated areas were 19.88 and 6.06 for

the unwashed and 5-times-washing samples, respectively. This indicates a residual fraction of 9.38% and an equivalent SDS concentration of approximately 0.94 mg mL^{-1} .

The two independent SDS-associated bands therefore indicate that approximately 3-10% of the initial SDS remained associated with the washed nanoparticle solids after five centrifugation cycles. Because this analysis relies on FTIR band-area ratios in a multicomponent solid matrix and may be affected by peak overlap, these values give a semi-quantitative estimation of residual SDS. Nevertheless, both spectral markers consistently show that the washing procedure removed most of the SDS, while a small residual fraction remained.

Supplementary Note 4 | Energy loss analysis

The energy loss in organic solar cells is determined as:

$$\begin{aligned}
 E_{loss} &= E_g - qV_{oc} \\
 &= (E_g - qV_{oc}^{SQ}) + (qV_{oc}^{SQ} - qV_{oc}^{rad}) + (qV_{oc}^{rad} - qV_{oc}) \\
 &= \Delta E_1 + \Delta E_2 + \Delta E_3
 \end{aligned} \tag{5}$$

V_{oc}^{SQ} in the equation is the maximum voltage based on the Shockley-Queisser limit, which can be as:

$$V_{oc}^{SQ} = \frac{kT}{q} \ln\left(\frac{J_{sc}^{SQ}}{J_0^{SQ}} + 1\right) \cong \frac{kT}{q} \ln\left(\frac{q \int_{E_g}^{\infty} \phi_{AM1.5}(E) dE}{q \int_{E_g}^{\infty} \phi_{BB}(E) dE} + 1\right) \tag{6}$$

When EQE_{EL} is unity, all the recombination is radiative, resulting in radiative limit V_{oc}^{rad} .

$$V_{oc}^{rad} = \frac{kT}{q} \ln\left(\frac{J_{sc}}{J_0^{rad}} + 1\right) = \frac{kT}{q} \ln\left(\frac{q \int_0^{\infty} EQE_{PV}(E) \cdot \phi_{AM1.5}(E) dE}{q \int_0^{\infty} EQE_{PV}(E) \cdot \phi_{BB}(E) dE} + 1\right) \tag{7}$$

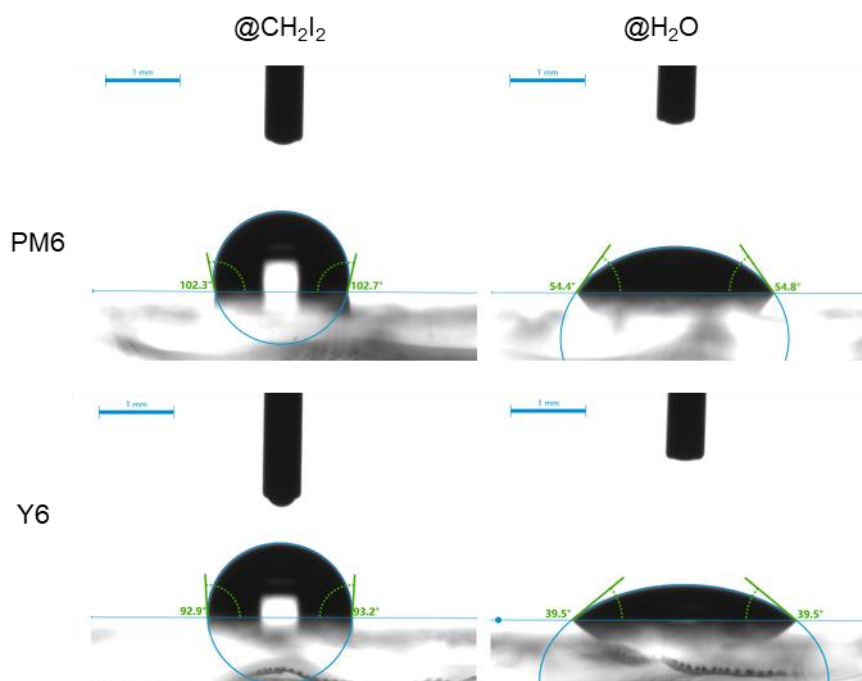
In this study, we considered the blend E_g as the derivatives of the EQE curves and can be determined as:

$$E_g = \frac{\int_a^b E_g \cdot P(E_g) dE}{\int_a^b P(E_g) dE} \tag{8}$$

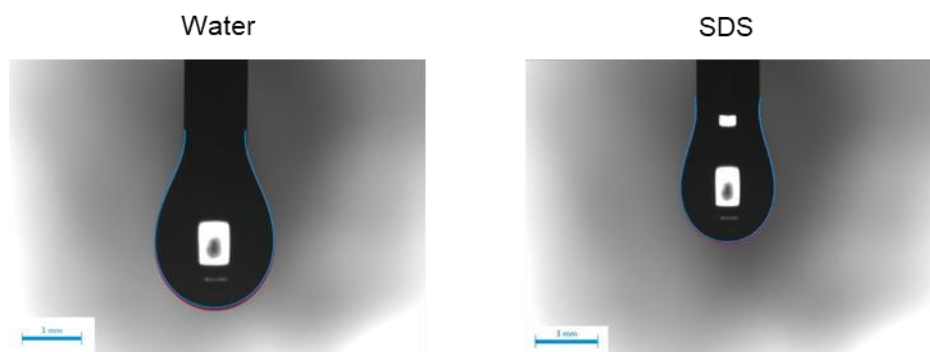
where the integral boundaries a and b are selected as $P(a) = P(b) = 0.5 \max P(E_g)$.

The detailed voltage losses were summarized in Supplementary Table 24.

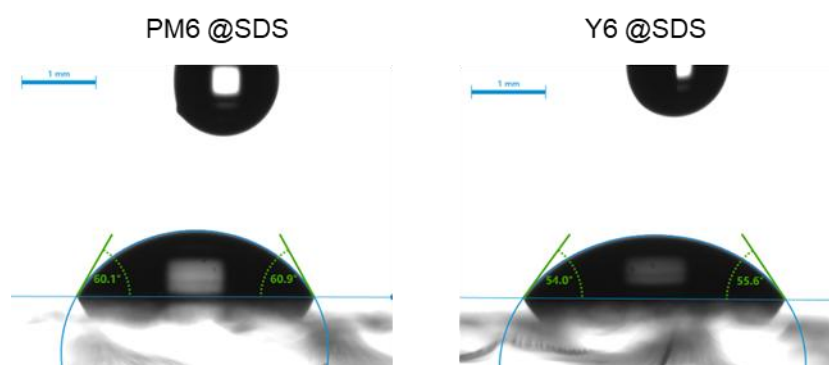
Supplementary Figures



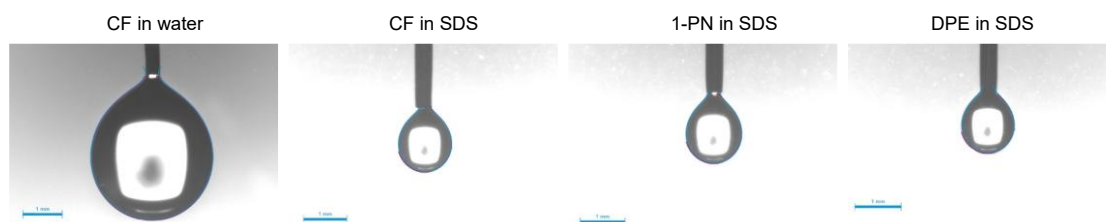
Supplementary Fig. 2 | Static contact angle measurement. Static contact angles of water and diiodomethane (CH_2I_2) on pristine PM6 and Y6 films.



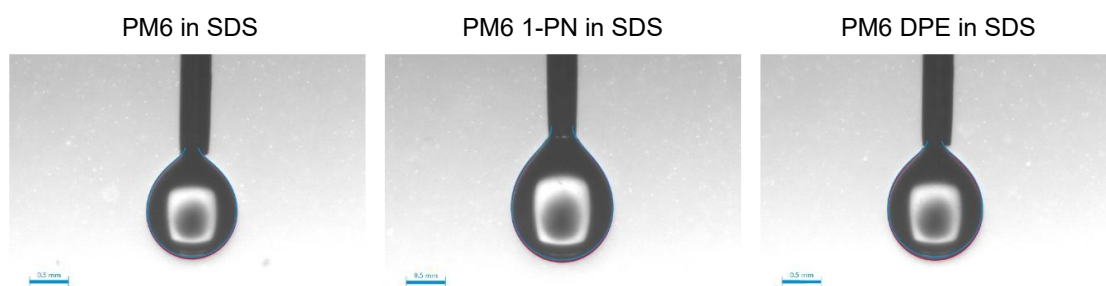
Supplementary Fig. 3 | Pendant-drop surface tension measurement. Surface tension of deionized water and SDS aqueous solution measured in air.



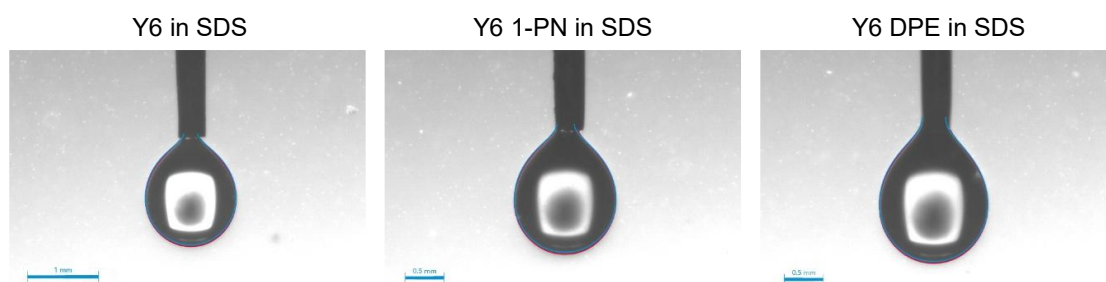
Supplementary Fig. 4 | Static contact angle measurement. Static contact angles of SDS aqueous solution on pristine PM6 and Y6 films.



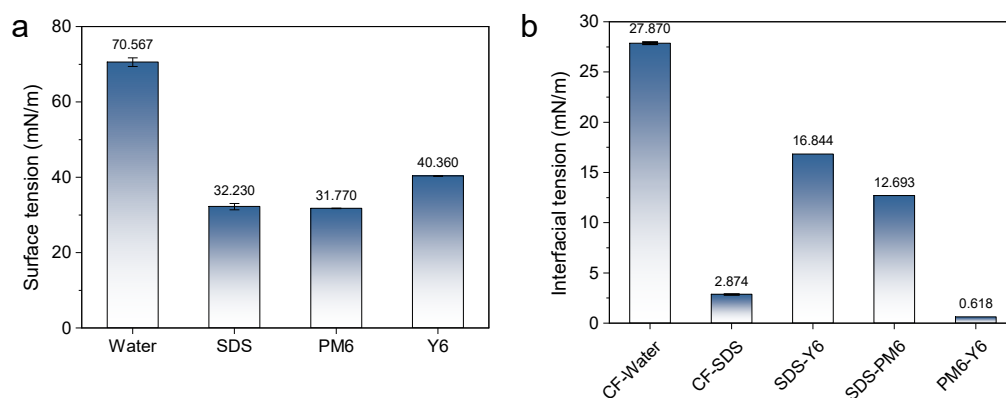
Supplementary Fig. 5 | Liquid-liquid interfacial tension measurements. Pendant-drop interfacial tension measurement of CF without and with additives (1.5 vol%) in water and SDS aqueous solution, respectively.



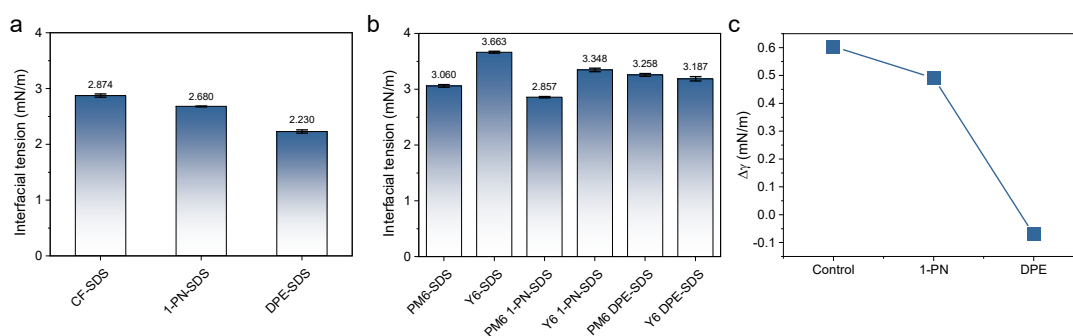
Supplementary Fig. 6 | Liquid-liquid interfacial tension measurements. Pendant-drop interfacial tension measurement of PM6 solution (0.5 mg/ml in CF) without and with additives (1.5 vol%) in SDS aqueous solution.



Supplementary Fig. 7 | Liquid-liquid interfacial tension measurements. Pendant-drop interfacial tension measurement of Y6 solution (0.5 mg/ml in CF) without and with additives (1.5 vol%) in SDS aqueous solution.

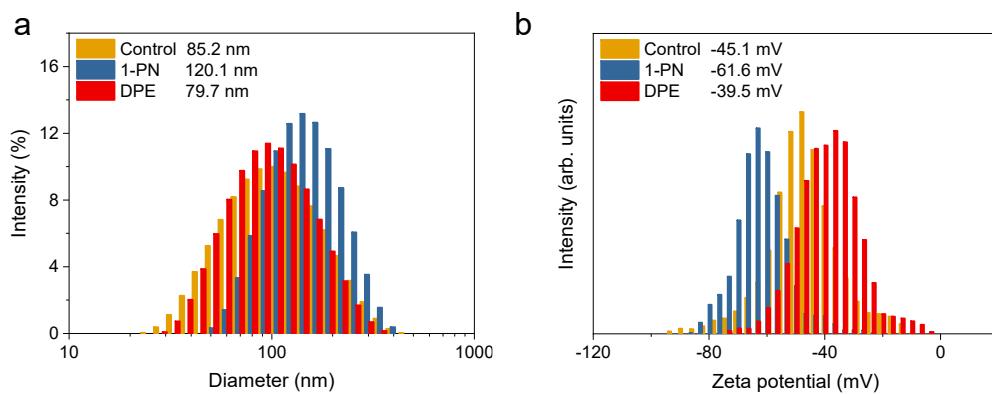


Supplementary Fig. 8 | Surface energy and interfacial tension analysis. a, Surface tension (or surface free energy for solids) of water, SDS solution, PM6 and Y6. **b,** Interfacial tension for chloroform (CF)-water, CF-SDS, Y6-SDS, PM6-SDS, and PM6-Y6, respectively. For the directly measured surface and interfacial tensions, error bars represent the temporal standard deviations over the stable acquisition interval.



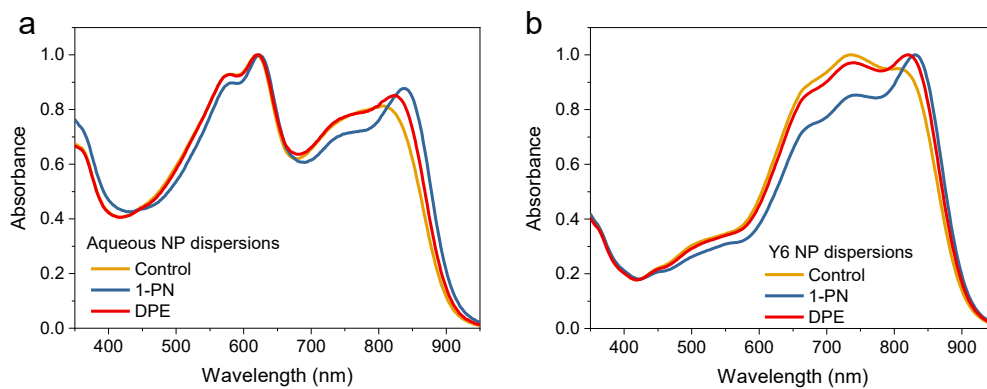
Supplementary Fig. 9 | Liquid-liquid interfacial tension measurements. a,

Interfacial tension between CF (with and without additives) and SDS aqueous solution. **b**, Interfacial tension between CF containing PM6 or Y6 (with and without additives) and SDS aqueous solution. **c**, Additive-dependent differences in liquid-liquid interfacial tension between PM6:Y6 chloroform solutions and SDS aqueous solution. Organic-aqueous interfacial tensions measured using the pendant-drop method. PM6 and Y6 were dissolved in the chloroform phase, SDS was dissolved in the aqueous phase, and additives (1-PN and DPE) were introduced in the chloroform phase. For the directly measured surface and interfacial tensions, error bars represent the temporal standard deviations over the stable acquisition interval.



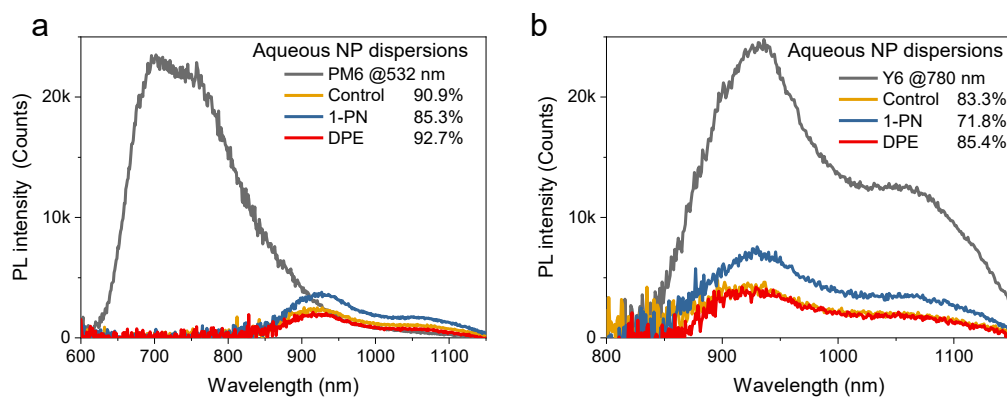
Supplementary Fig. 10 | DLS size distribution and zeta-potential measurements.

a, DLS size distribution of PM6:Y6 NP dispersions after storage at room temperature for 10 days. b, Zeta-potentials of the PM6:Y6 NP dispersions.

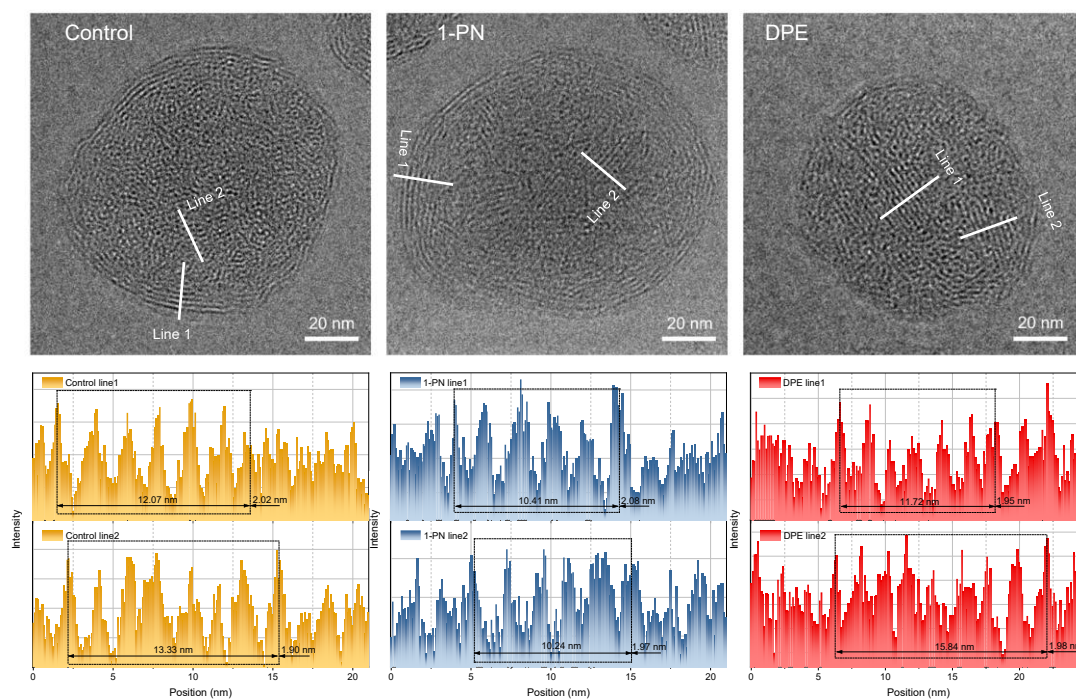


Supplementary Fig. 11 | UV-vis absorption of aqueous NP dispersions.

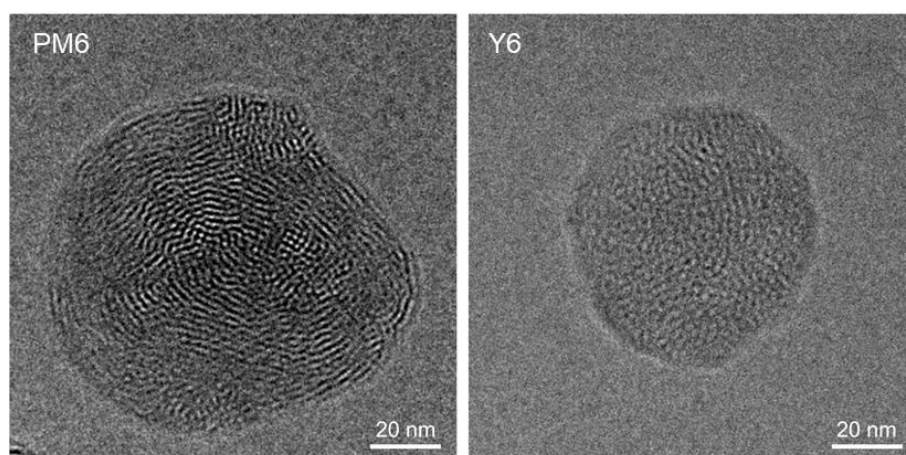
Normalized UV-vis absorption spectra of aqueous PM6:Y6 NP dispersions (**a**) and neat Y6 NP dispersions (**b**) prepared without additives (control) and with non-halogenated solvent additives (1-PN or DPE).



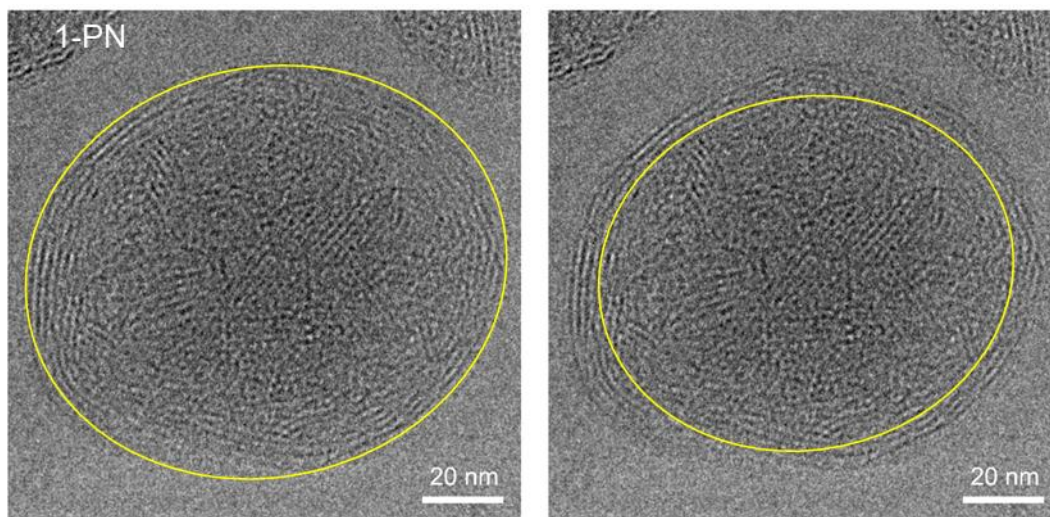
Supplementary Fig. 12 | Photoluminescence of PM6:Y6 NP dispersions. PL spectra of aqueous PM6:Y6 NP dispersions excited at 532 nm (**a**) and 780 nm (**b**).



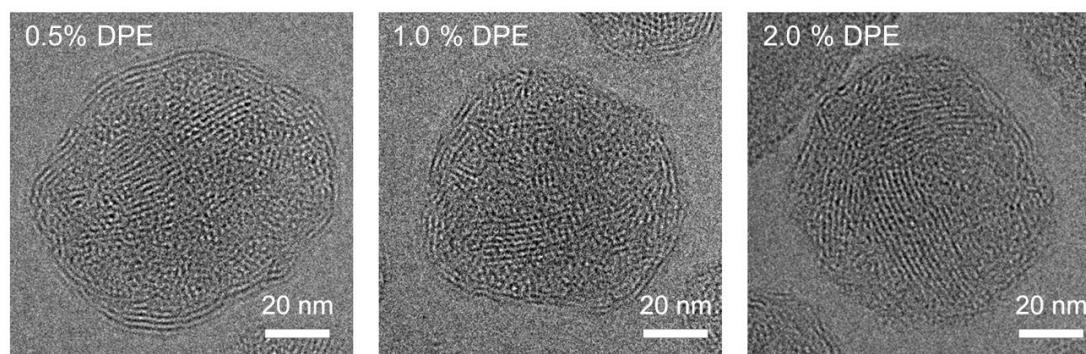
Supplementary Fig. 13 | Cryo-TEM analysis of PM6:Y6 NPs. High resolution cryo-TEM images of PM6:Y6 blend NPs prepared without additives (control) and with non-halogenated solvent additives (1-PN or DPE). Intensity profiles along the marked lines and the corresponding lattice spacings are shown.



Supplementary Fig. 14 | Cryo-TEM image of neat PM6 and Y6 NPs. PM6 NP exhibits continuous, curved elongated textures, whereas Y6 NP shows locally ordered polycrystalline features.

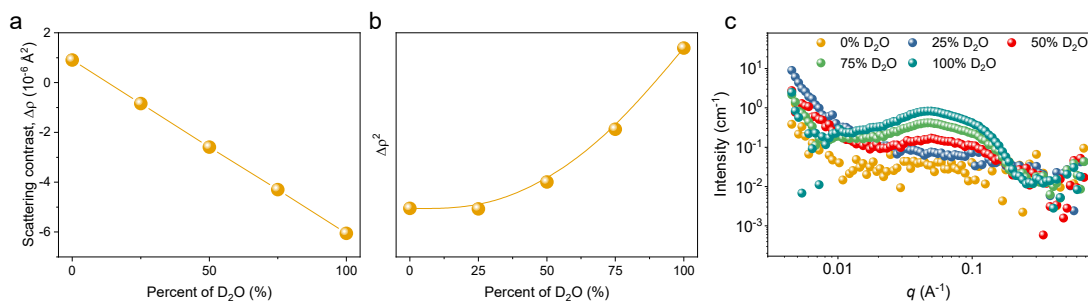


Supplementary Fig. 15 | Geometric mass-balance analysis for 1-PN processed PM6:Y6 NPs. Volume estimates assuming a prolate-ellipsoidal particle geometry and an equivalent uniform shell thickness derived from the measured long/short axes. Because 1-PN-processed PM6:Y6 NPs show an ellipsoidal projection, we approximate the 3D morphology as a prolate ellipsoid with semi-axes $a = 58$ and $b = c = 49$ nm, and represent the shell by an equivalent uniform thickness of 8 nm (noting local non-uniformity and discontinuities). This geometry yields a shell fraction of about 40 vol% for the representative particle. Combined with the global PM6:Y6 = 1:1.2 (w/w) formulation and bulk densities, the resulting mass balance cannot be satisfied by assigning a phase-pure PM6 shell and confining Y6 to the core, supporting partial intermixing and/or a graded composition across the shell-core regions rather than strictly phase-pure domains.

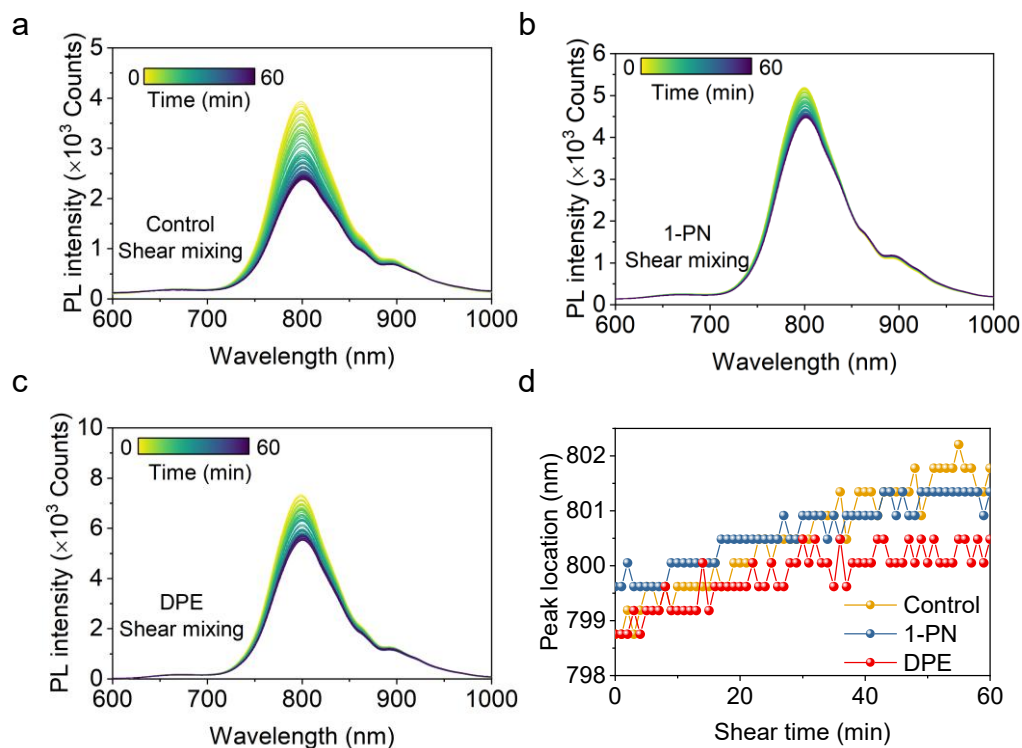


Supplementary Fig. 16 | Cryo-TEM image of DPE processed PM6:Y6 NPs.

Representative images of PM6:Y6 NPs prepared with different DPE volume fractions.

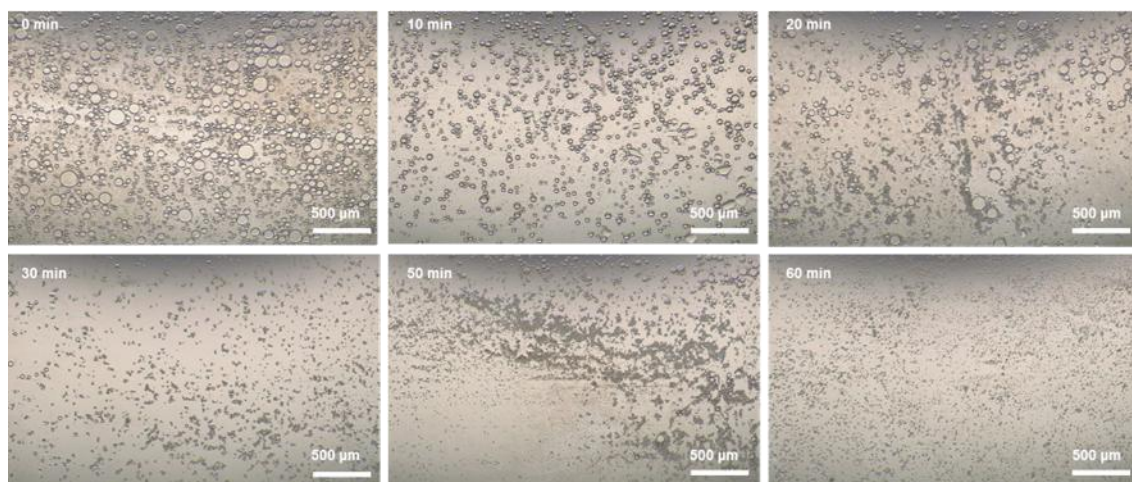


Supplementary Fig. 17 | SANS analysis of SDS aqueous solutions. a, Scattering contrast ($\Delta\rho$) between SDS and solvents in different $\text{H}_2\text{O}:\text{D}_2\text{O}$ mixtures. SLD of H_2O , D_2O , and SDS are -0.56×10^{-6} , 6.4×10^{-6} , and $0.34 \times 10^{-6} \text{ \AA}^{-2}$ respectively. **b,** Variation of $\Delta\rho^2$ between SDS and solvents with the increasing ratio of D_2O in the $\text{H}_2\text{O}:\text{D}_2\text{O}$ mixtures. **c,** SANS profiles of SDS aqueous solutions measured in different $\text{H}_2\text{O}:\text{D}_2\text{O}$ mixtures.

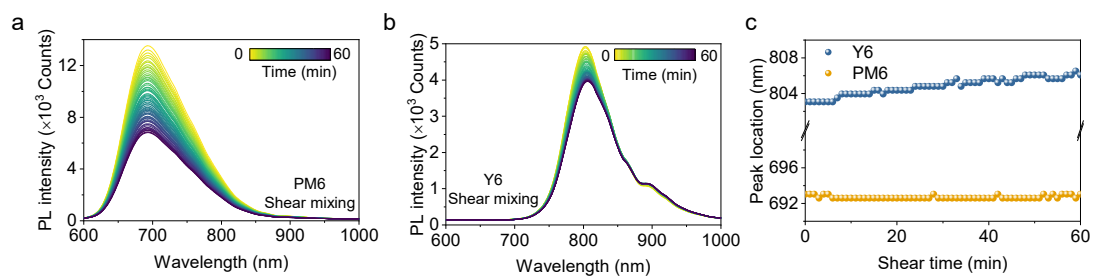


Supplementary Fig. 18 | In-situ PL during shear mixing of PM6:Y6 NP

dispersions. a-c, Evolution of in situ PL spectra during shear mixing of water/chloroform emulsions. **d,** Time evolution of the PL peak position for PM6:Y6 dispersions prepared with and without additive during the shear-mixing stage.



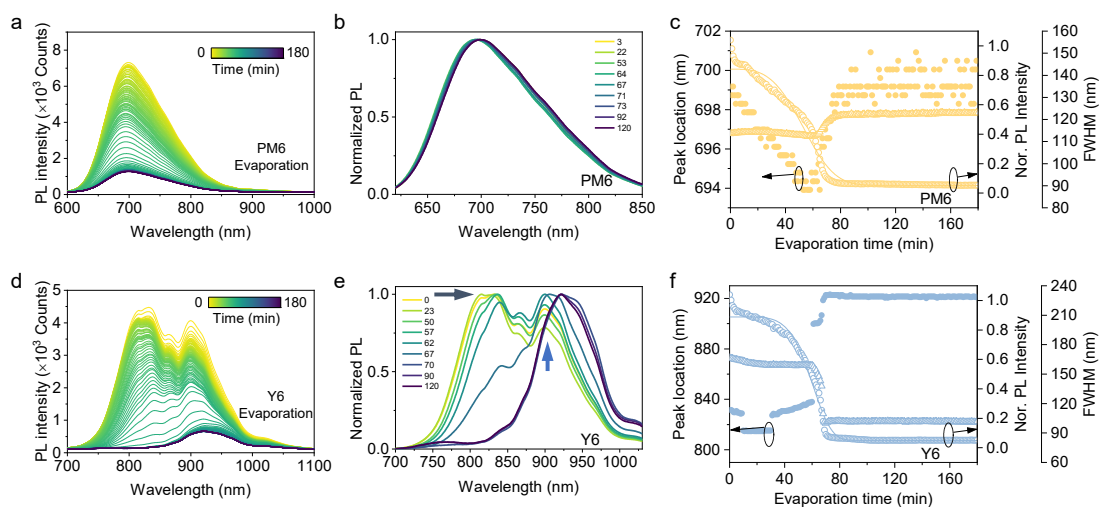
Supplementary Fig. 19 | Optical microscope during shear mixing. Optical micrographs of emulsion droplets collected at different shear-mixing times for CF droplets in water/SDS emulsion (SDS concentration: 10 mg ml⁻¹).



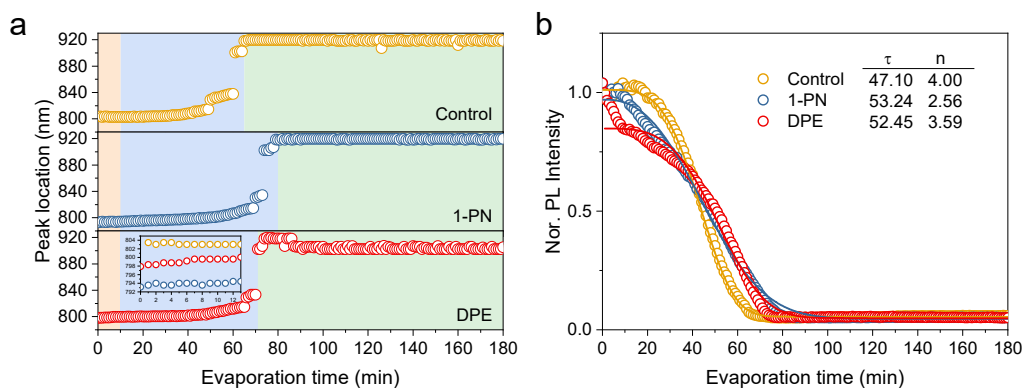
Supplementary Fig. 20 | In-situ PL during shear mixing of neat NP dispersions.

a-b, In-situ PL spectra during water/CF mixing for neat PM6 and Y6 NP dispersions.

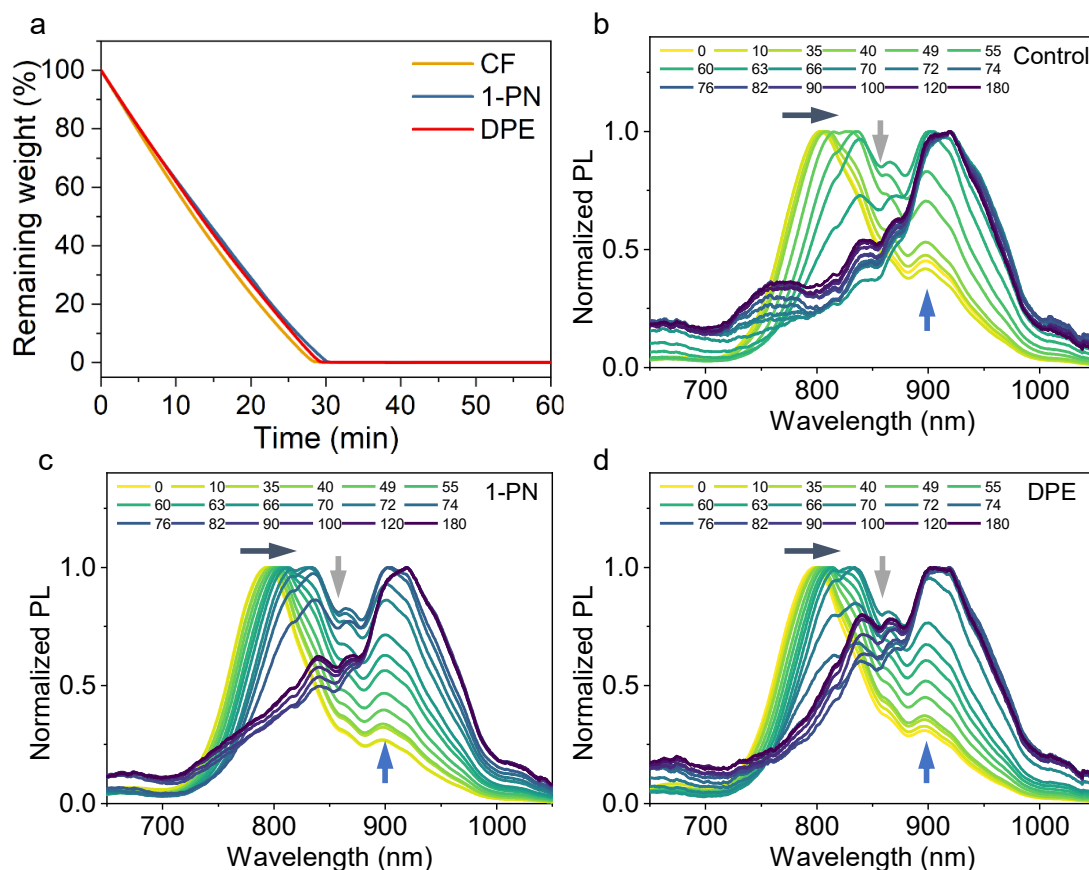
c, Time evolution of the PL peak position during the shear-mixing stage.



Supplementary Fig. 21 | In-situ PL during CF evaporation for neat NP dispersions. **a-c**, Evolution of in situ PL spectra during CF evaporation (**a**), normalized PL spectra measured at different minute time points (**b**), and time evolution of emission- peak position, intensity, and full width at half maximum (FWHM) during NP formation (**c**) for neat PM6 NP dispersion. **d-f**, Evolution of in situ PL spectra during CF evaporation (**d**), normalized PL spectra measured at different minute time points (**e**), and time evolution of emission- peak position, intensity, and FWHM during NP formation (**f**) for neat Y6 NP dispersion.

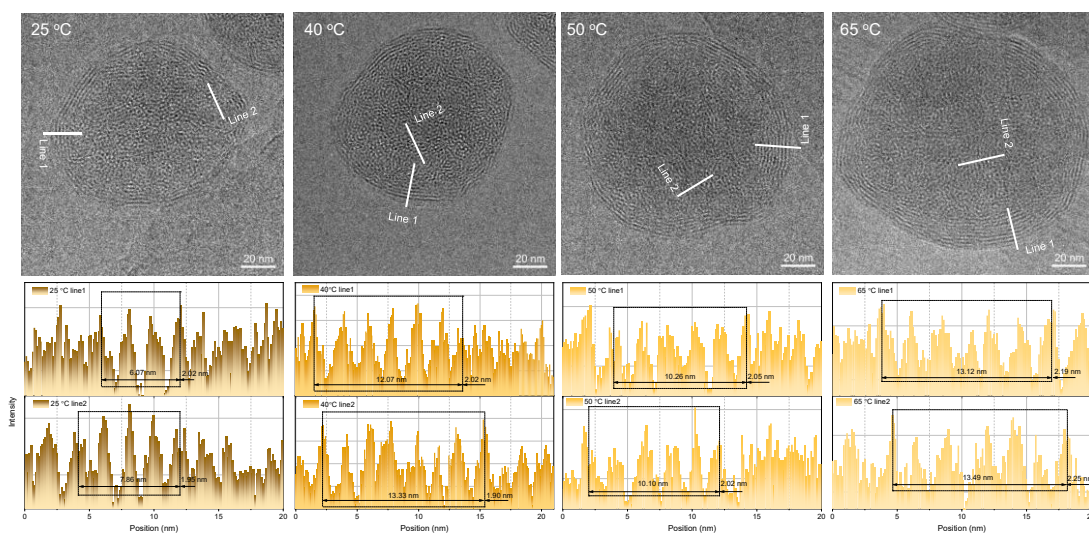


Supplementary Fig. 22 | In-situ PL measurements during CF evaporation for PM6:Y6 NP dispersions. Time evolution of PL peak position (a) and PL peak intensity (b) for PM6:Y6 dispersions prepared with and without additive during CF evaporation. Inset in panel a shows an expanded view of the first 12 min, highlighting that the PL peak in the DPE condition begins to red-shift within the first 10 min, whereas the control and 1-PN conditions remain nearly unchanged over the same interval. Panel b shows Kohlrausch-Williams-Watts (KWW) fits (lines)² to the PL-intensity evolution, yielding the characteristic time constants (τ) and stretching exponents (n).

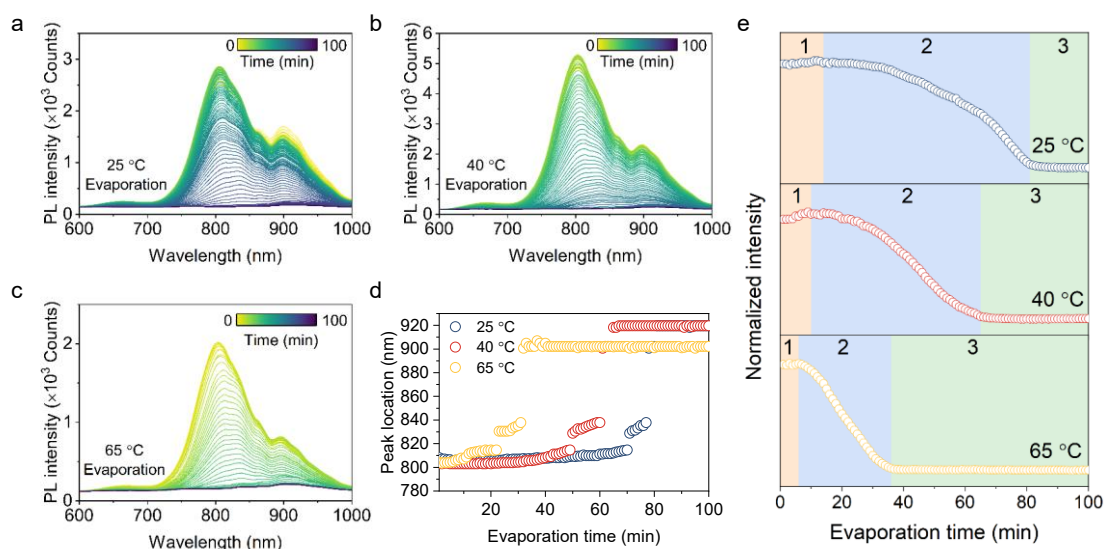


Supplementary Fig. 23 | TGA evaporation measurement and normalized in-situ PL spectra analysis. **a**, TGA evaporation curves denoting the remaining weight over time for a 15 μ l droplet of each emulsion. **b-d**, The normalized PL spectra of PM6:Y6 dispersions without additives (**b**) and with 1-PN (**c**) or DPE (**d**) measured at different minute time points during the evaporation process.

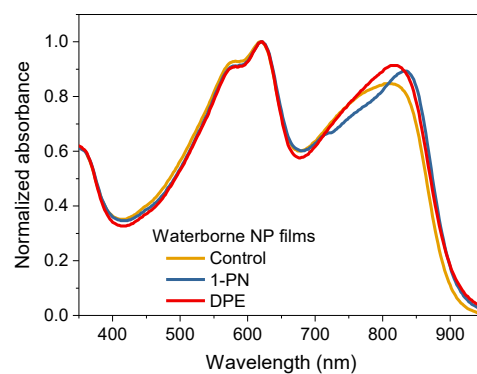
The contribution of the low-energy Y6 emission near 900 nm increased and became pronounced at approximately 55 min in the additive-treated emulsions, consistent with the later development of more strongly coupled Y6 aggregates. In the control sample, however, this spectral change occurred earlier and overlapped with the initial intensity decrease from about 40 min, resulting in a more continuous PL decay. We therefore attribute the two-step behavior primarily to the temporal separation between initial densification-driven quenching and the subsequent evolution of the low-energy Y6 aggregate state.



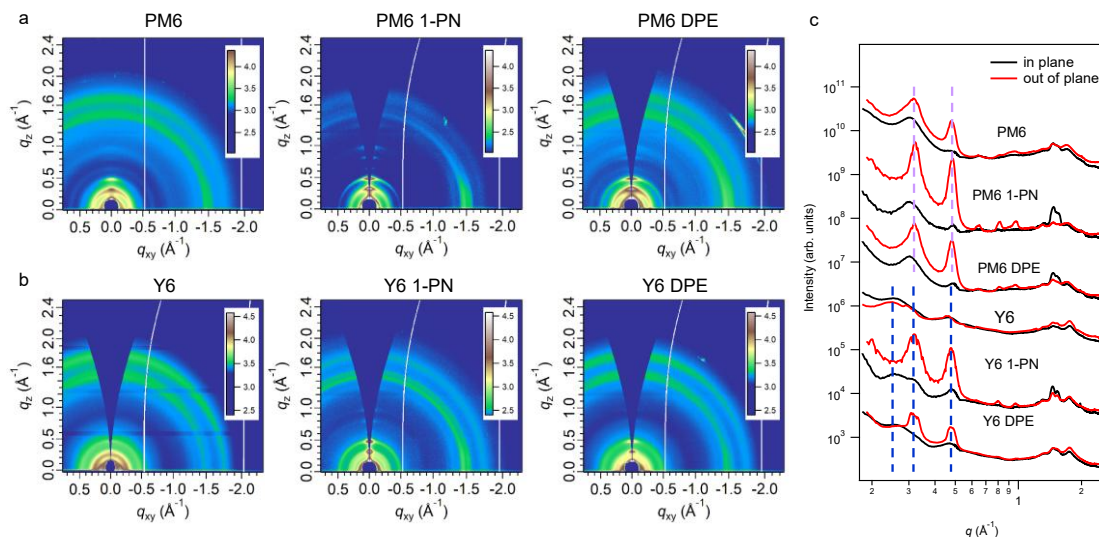
Supplementary Fig. 24 | Cryo-TEM analysis of PM6:Y6 NPs prepared at different evaporation temperatures. High resolution cryo-TEM images of PM6:Y6 NPs prepared at different evaporation temperatures (25 °C, 40 °C, 50 °C, and 65 °C). Intensity profiles along the marked lines and the corresponding lattice spacings are shown.



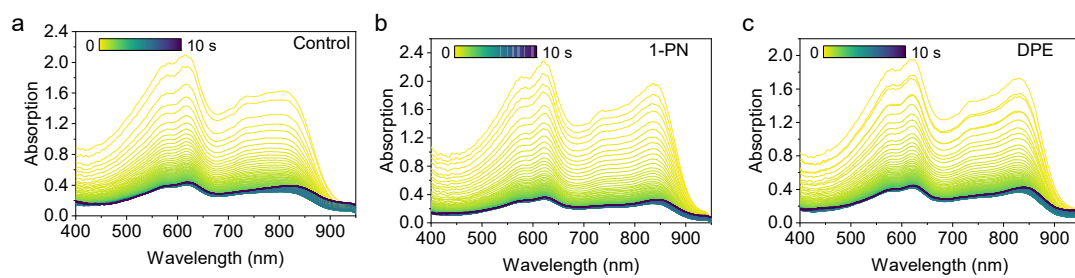
Supplementary Fig. 25 | In situ PL analysis of PM6:Y6 NPs prepared at different evaporation temperatures. (a-c) Time-resolved PL spectra during evaporation for PM6:Y6 dispersions with different evaporation temperature. Time evolution of PL peak position (d) and PL peak intensity (e) for PM6:Y6 dispersions prepared at different evaporation temperature.



Supplementary Fig. 26 | UV-vis Absorption measurements of waterborne PM6:Y6 NP films. Normalized UV-vis absorption spectra of waterborne PM6:Y6 NP films prepared without and with non-halogenated solvent additives.

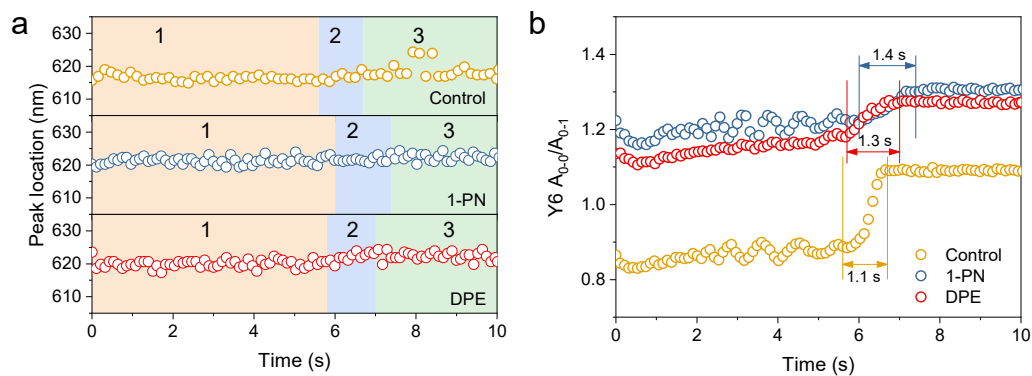


Supplementary Fig. 27 | GIWAXS data of neat PM6 and neat Y6 films. Two-dimensional GIWAXS patterns (a, b) and corresponding linecuts (c) of neat PM6 and neat Y6 NP-derived films deposited from concentrated NP inks without and with additives.

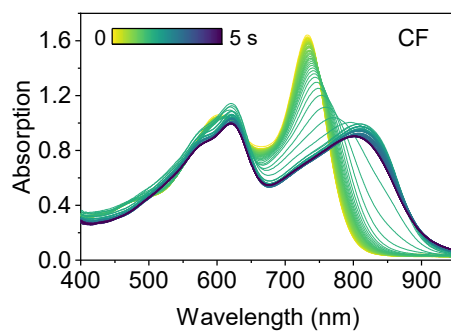


Supplementary Fig. 28 | In situ UV-vis absorption during NP-film deposition.

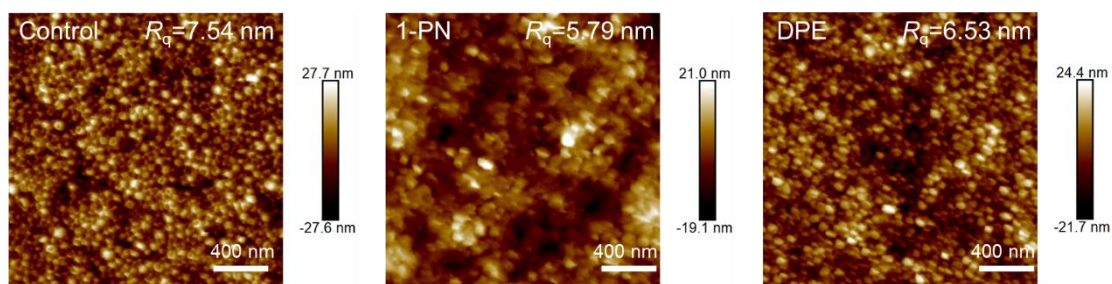
Time-resolved UV-vis absorption spectra recorded during NP-film formation from aqueous NP inks for control, 1-PN and DPE conditions.



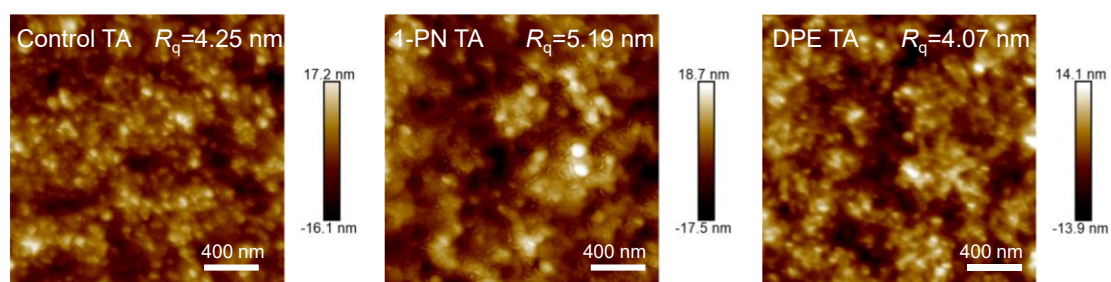
Supplementary Fig. 29 | In situ UV-vis analysis during NP-film deposition. a, Time evolution of the PM6 absorption peak position during NP-film formation from aqueous NP inks for control, 1-PN and DPE conditions. **b,** Time evolution of the Y6 A_{0-0}/A_{0-1} (approximated as A_{844}/A_{750}) vibronic peak intensity ratio during nanoparticle-based film formation for control, DPE, and 1-PN NPs.



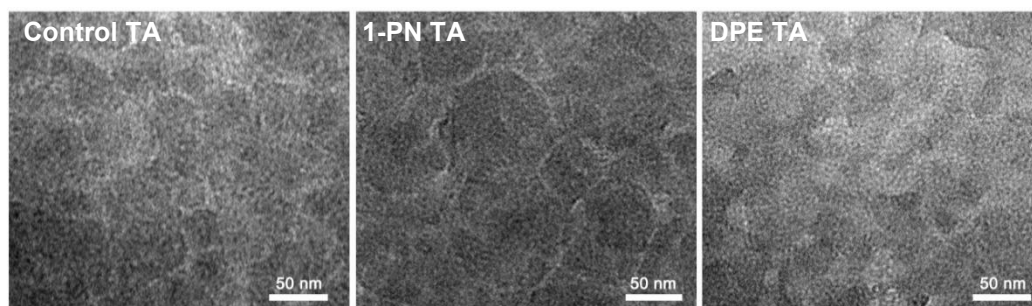
Supplementary Fig. 30 | In situ UV-vis measurement during spin-coating from chloroform solution. Time-resolved UV-vis absorption spectra recorded during spin-coating of PM6:Y6 films from chloroform solution.



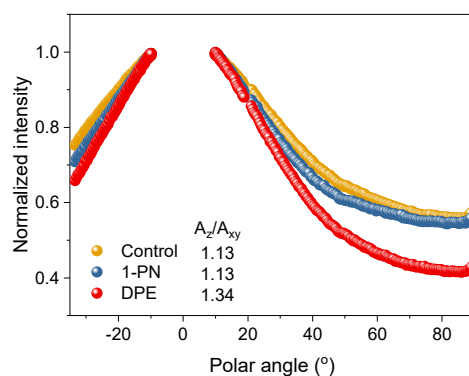
Supplementary Fig. 31 | AFM height images of as-cast NP films. AFM height images of as-cast PM6:Y6 NP films prepared without additives (control) and with non-halogenated solvent additives (1-PN or DPE).



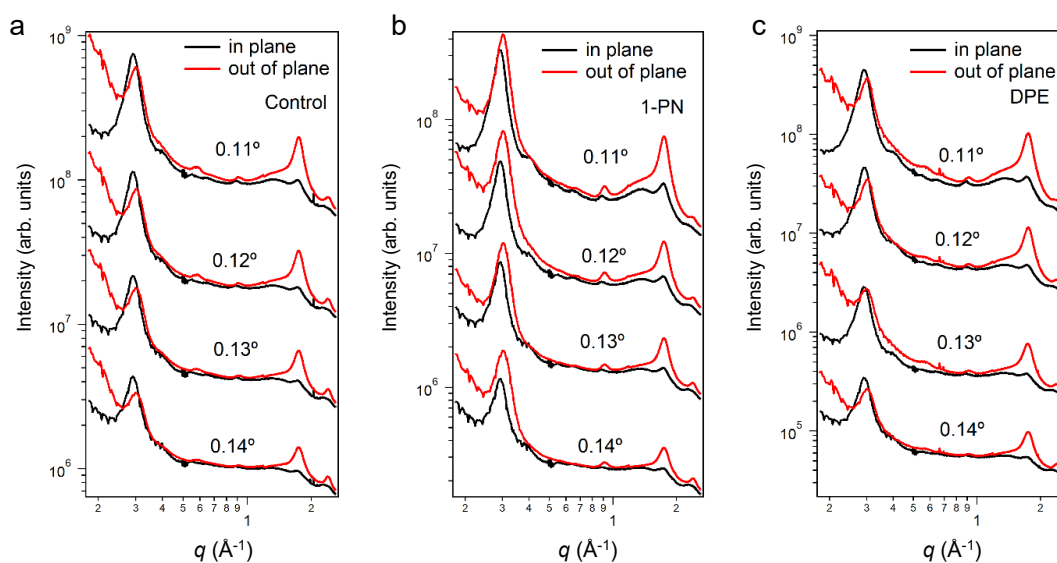
Supplementary Fig. 32 | AFM height images after thermal annealing. AFM height images of thermally annealed PM6:Y6 NP films prepared without additives (control) and with non-halogenated solvent additives (1-PN or DPE).



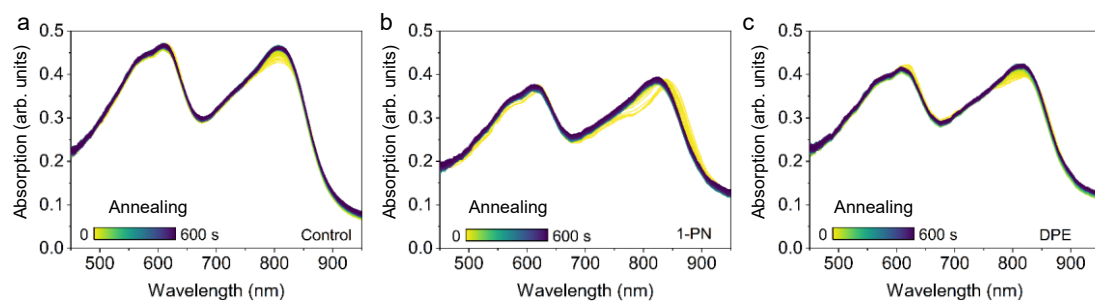
Supplementary Fig. 33 | TEM images after thermal annealing. TEM images of thermally annealed PM6:Y6 NP films prepared without additives (control) and with solvent additives (1-PN or DPE).



Supplementary Fig. 34 | GIWAXS pole figures of thermally annealed PM6:Y6 NP films. Normalized pole figures extracted from the (010) diffraction peak for thermally annealed PM6:Y6 NP films prepared without additives (control) and with non-halogenated solvent additives (1-PN or DPE).

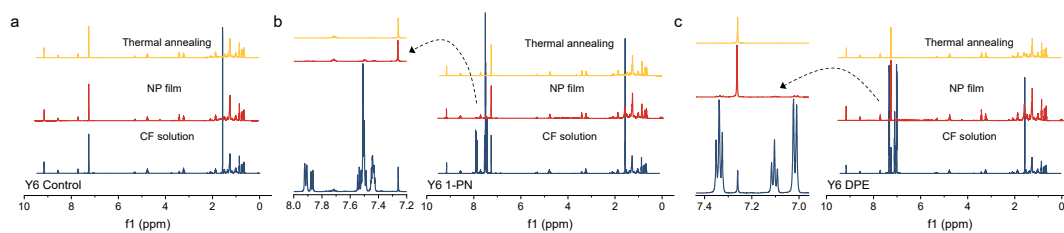


Supplementary Fig. 35 | Incident-angle-dependent GIWAXS line cuts. In-plane and out-of-plane GIWAXS line-cut profiles of thermally annealed PM6:Y6 NP films prepared without additives (control) and with solvent additives (1-PN or DPE), measured at incident angles of 0.11-0.14°.

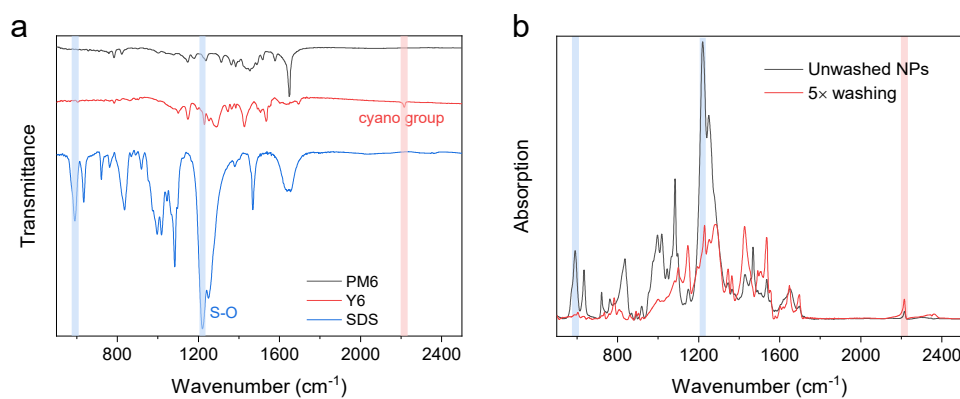


Supplementary Fig. 36 | In situ UV-vis absorption during thermal annealing.

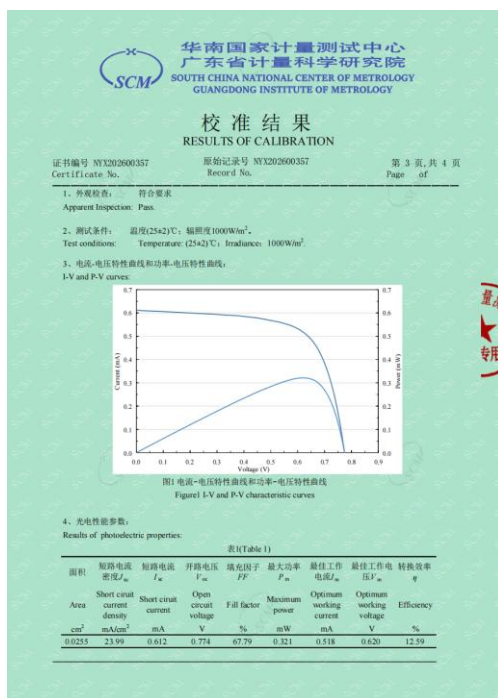
Time-resolved UV-vis absorption spectra of PM6:Y6 NP films during thermal annealing for control (a), 1-PN (b) and DPE (c) conditions.



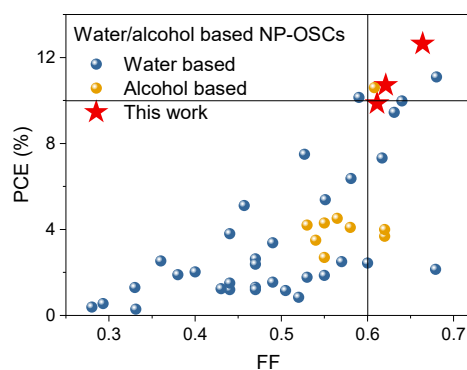
Supplementary Fig. 37 | Characterization of residual additives. NMR ^1H spectra of Y6 samples without and with additives in CDCl_3 .



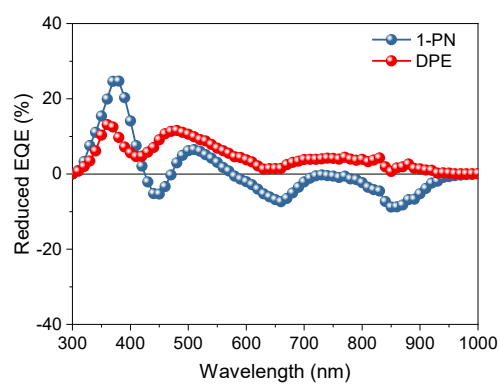
Supplementary Fig. 38 | Characterization of residual SDS. Fourier-transform infrared spectra of neat PM6, neat Y6, and neat SDS (a) and PM6:Y6·NPs before and after washing process (b).



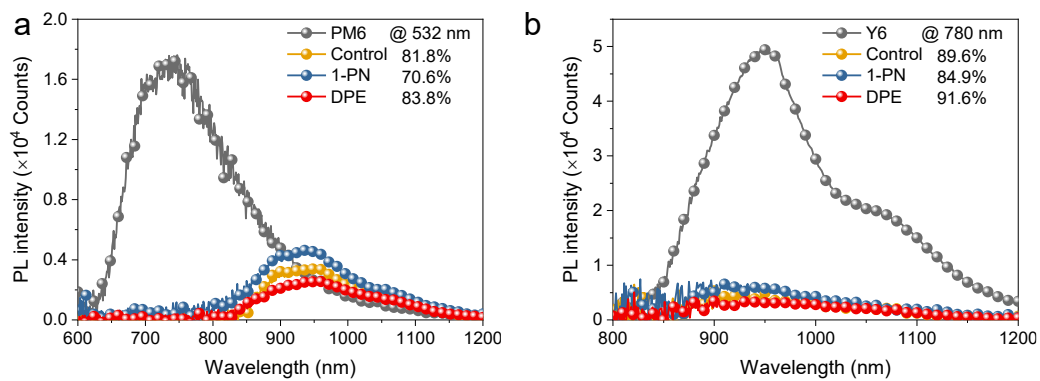
Supplementary Fig. 39 | Certification report from South China National Center of Metrology Guangdong Institute of Metrology.



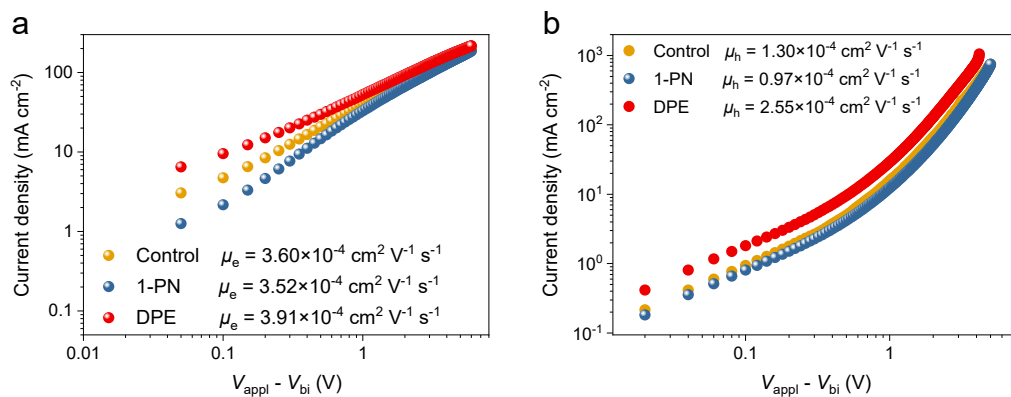
Supplementary Fig. 40 | Literature comparison of PCE versus FF for water/alcohol-based NP-OSCs. Detailed data are presented in Supplementary Table19.



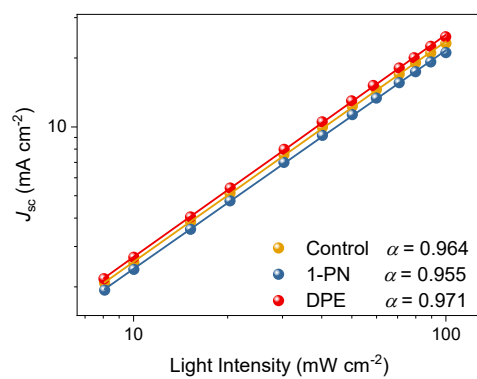
Supplementary Fig. 41 | Reduced EQE spectra. Reduced EQE spectra of the 1-PN and DPE devices relative to the control device.



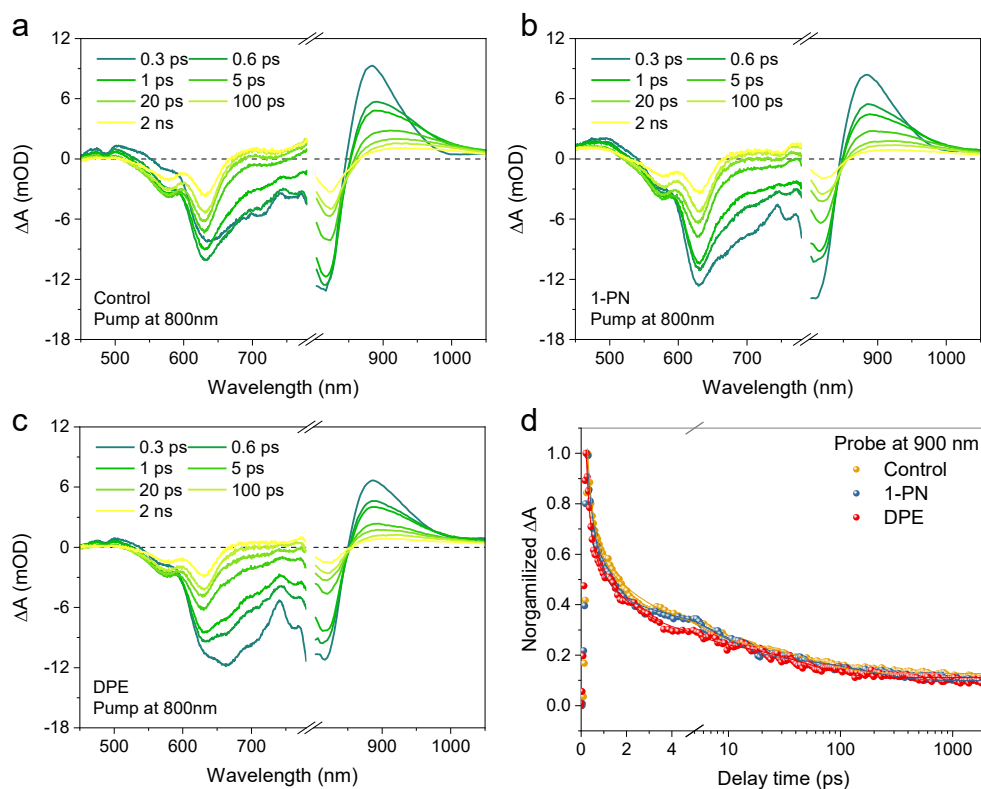
Supplementary Fig. 42 | PL spectra of NP films. a, PL spectra of PM6 and PM6:Y6 NP films excited at 532 nm. **b,** PL spectra of the neat Y6 NP film and blend NP films excited at 780 nm.



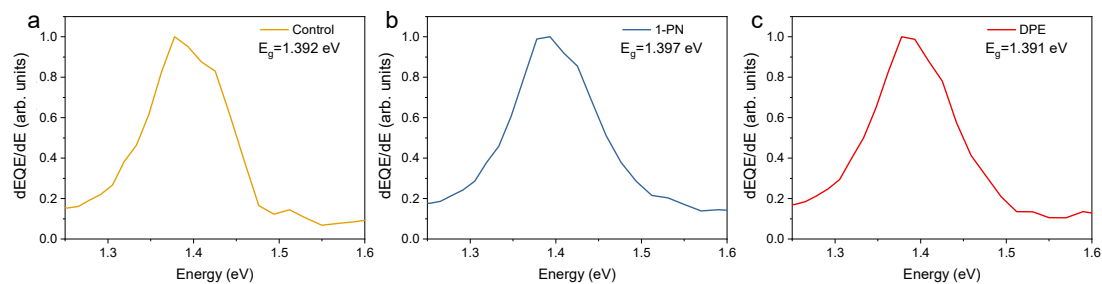
Supplementary Fig. 43 | SCLC mobility measurements. Dark J - V characteristics of electron-only devices (**a**) and hole only devices (**b**) for waterborne PM6:Y6 NP devices prepared under control, 1-PN and DPE conditions.



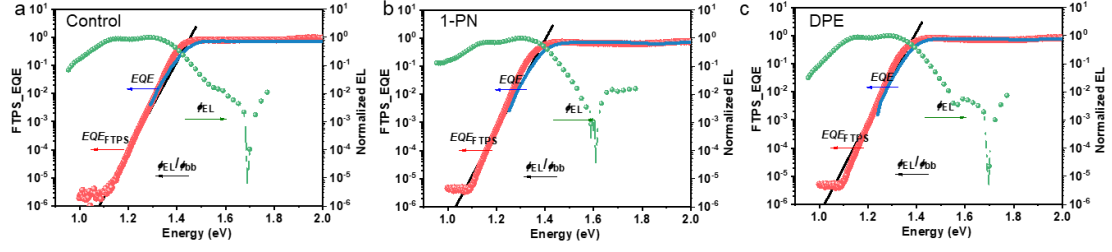
Supplementary Fig. 44 | Charge recombination analysis. Plots of J_{sc} versus incident light intensity of devices prepared under control, 1-PN and DPE conditions.



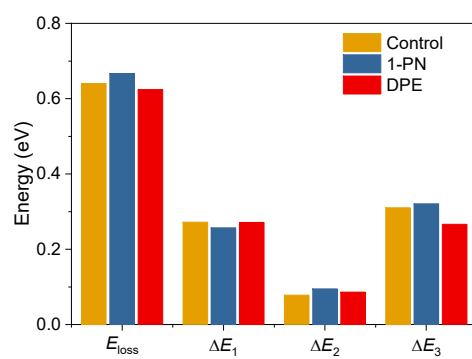
Supplementary Fig. 45 | Transient absorption spectroscopy. a-c, TA spectra of thermally annealed waterborne PM6:Y6 NP films recorded at different pump-probe delay times. **d,** Kinetic traces of the Y6 LE signals at 900 nm (symbols) and related fits (lines).



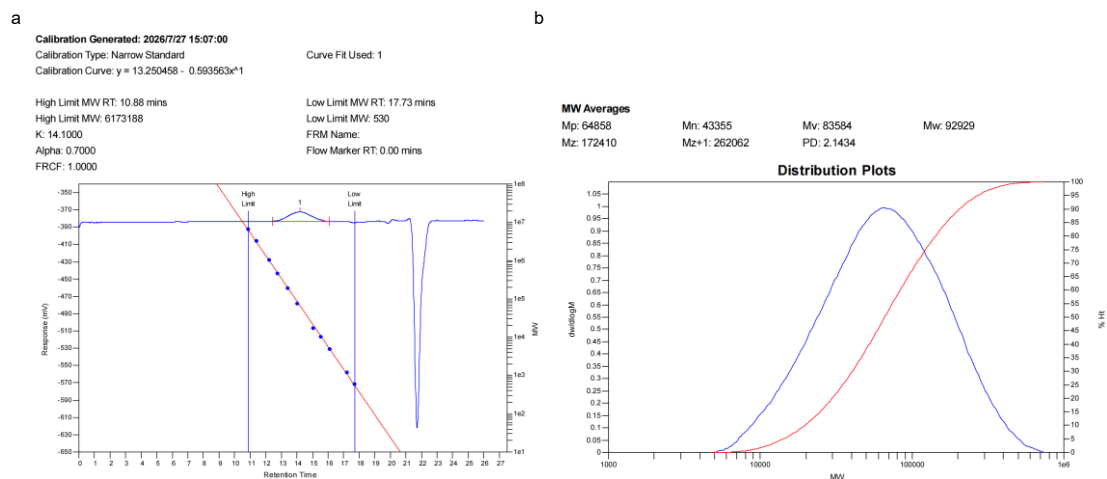
Supplementary Fig. 46 | Bandgap analysis. a-c, Bandgap analysis for PM6:Y6 NP devices prepared under control, 1-PN and DPE conditions. The E_g value is determined by the derivatives of the FTPS-EQE curve and then calculated according to the literature.



Supplementary Fig. 47 | FTPS-EQE and electroluminescence (EL) analysis. a-c, Semilogarithmic plots of normalized EL, measured EQE, and FTPS-EQE spectra as a function of energy for PM6:Y6 NP devices prepared under control, 1-PN and DPE conditions. The ratio of $\phi_{\text{EL}}/\phi_{\text{bb}}$ was used to plot the EQE in the low-energy regime, where ϕ_{EL} and ϕ_{bb} represent the emitted photon flux and the room-temperature blackbody photon flux, respectively.



Supplementary Fig. 48 | Voltage loss analysis. Statistical diagram of energy loss for the PM6:Y6 NP devices prepared under control, 1-PN and DPE conditions.



Supplementary Fig. 49 | Molecular weight characterization. Gel-permeation chromatography trace of PM6.

Supplementary Tables

Supplementary Table 1 | Surface free energy of PM6 and Y6 films.

	γ (mN m ⁻¹)	γ^d (mN m ⁻¹)	γ^p (mN m ⁻¹)
PM6	31.77	31.68	0.09
Y6	40.36	39.91	0.46

Supplementary Table 2 | Static contact angles of water and SDS solution on PM6 and Y6 films and the corresponding estimated interfacial tensions.

	Contact angle (°)		Interfacial tension (mN m ⁻¹)	
	θ^{water}	θ^{SDS}	water	SDS
PM6	102.65	53.71	47.224	12.693
Y6	93.27	43.14	44.385	16.844

Supplementary Table 3 | Vibronic intensity ratio A_{0-0}/A_{0-1} in aqueous NP dispersions

	PM6:Y6 NP dispersions ^a	neat Y6 NP dispersions ^b
Control	1.07	0.95
1-PN	1.25	1.17
DPE	1.12	1.03

^a The Vibronic intensity ratio A_{0-0}/A_{0-1} was estimated by using the absorption peak intensity of NP dispersions at 806-836 nm and 740 nm.

^b The Vibronic intensity ratio A_{0-0}/A_{0-1} was estimated by using the absorption peak intensity of NP dispersions at 810-832 nm and 740 nm.

Supplementary Table 4 | The parameters and results of fitting the SANS profiles of PM6:*d*-Y6 NP dispersions in 25% D₂O water with the combination of fractal-like network model and core-shell model.

25% D ₂ O	afrac	rfrac (Å)	zfrac	b (cm ⁻¹)	ccfrac (Å)	ddfrac	<i>R_g</i> (nm)	thickness (nm)
Control	1.33×10 ⁻⁴	24.99	5.21	0.10	275.51	2.39	33.59	6.35
1-PN	2.99×10 ⁻⁴	33.19	4.57	0.10	262.21	2.82	35.15	8.33
DPE	3.31×10 ⁻⁴	10.01	23.75	0.11	113.19	2.95	27.57	--

Supplementary Table 5 | The parameters and results of fitting the SANS profiles of PM6:*d*-Y6 NP dispersions in 100% D₂O water with the fractal-like network model.

100% D ₂ O	afrac	rfrac (Å)	zfrac	b (cm ⁻¹)	Ccfrac (Å)	Ddfrac	<i>R_g</i> (nm)
Control	7.01×10 ⁻⁴	50.00	2.14	0.50	115.13	2.97	27.95
1-PN	3.34×10 ⁻⁴	93.33	0.81	0.81	168.20	2.78	38.55
DPE	1.24×10 ⁻⁴	130.05	1.16	1.30	132.30	2.35	26.23

Supplementary Table 6 | Out-of-plane (100) Peak parameters of as-cast PM6:Y6 NP films. Peak position q , d-spacing, FWHM and CCL.

OOP (100)	q ($\text{\AA}^{-1}$)	d -spacing ($\text{\AA}$)	FWHM ($\text{\AA}^{-1}$)	CCL ($\text{\AA}$)
Control	0.30	21.14	0.08	72.98
1-PN	0.32	19.59	0.05	124.92
	0.48	13.05	0.04	139.51
DPE	0.31	20.41	0.06	94.54
	0.48	13.14	0.08	67.65

Supplementary Table 7 | Out-of-plane (010) Peak parameters of as-cast PM6:Y6 NP films. Peak position q , d-spacing, FWHM and CCL.

OOP (010)	q ($\text{\AA}^{-1}$)	d -spacing ($\text{\AA}$)	FWHM ($\text{\AA}^{-1}$)	CCL ($\text{\AA}$)
Control	1.76	3.57	0.22	25.31
1-PN	1.74	3.61	0.20	27.86
DPE	1.75	3.59	0.21	26.84

Supplementary Table 8 | GIWAXS peak parameters of thermally annealed PM6:Y6 NP films. (100) and (010) peak position q , d-spacing, FWHM and CCL.

	q ($\text{\AA}^{-1}$)	d -spacing ($\text{\AA}$)	FWHM ($\text{\AA}^{-1}$)	CCL ($\text{\AA}$)
Control	0.29	21.59	0.04	128.68
	1.75	3.60	0.19	29.96
1-PN	0.29	21.50	0.04	136.79
	1.75	3.60	0.19	29.28
DPE	0.29	21.50	0.04	131.63
	1.75	3.59	0.19	29.64

Supplementary Table 9 | Relative fractions of face-on and edge-on orientations of thermally annealed PM6:Y6 NP films. Ratio of face-on (A_z) and edge-on (A_{xy}) contributions extracted from Fig. 4e.

	A_z (%)	A_{xy} (%)	A_z/ A_{xy}
Control	52.95	47.05	1.13
1-PN	52.95	47.05	1.13
DPE	57.33	42.67	1.34

Supplementary Table 10 | Incident-angle-dependent peak parameters for thermally annealed control NP films. (100) and (010) peak position q , d-spacing, FWHM and CCL at different incident angles.

Control	q ($\text{\AA}^{-1}$)	d -spacing ($\text{\AA}$)	FWHM ($\text{\AA}^{-1}$)	CCL ($\text{\AA}$)
0.11°	0.29	21.56	0.04	128.17
	1.75	3.60	0.19	30.25
0.12°	0.29	21.59	0.04	128.68
	1.75	3.60	0.19	29.96
0.13°	0.29	21.55	0.04	130.08
	1.75	3.60	0.19	29.36
0.14°	0.29	21.56	0.04	131.13
	1.75	3.60	0.19	29.34

Supplementary Table 11 | Incident-angle-dependent peak parameters for thermally annealed 1-PN NP films. (100) and (010) peak position q , d-spacing, FWHM and CCL at different incident angles.

1-PN	q ($\text{\AA}^{-1}$)	d -spacing ($\text{\AA}$)	FWHM ($\text{\AA}^{-1}$)	CCL ($\text{\AA}$)
0.11°	0.29	21.56	0.04	135.94
	1.75	3.60	0.19	29.58
0.12°	0.29	21.50	0.04	136.79
	1.75	3.60	0.19	29.28
0.13°	0.29	21.52	0.04	139.75
	1.75	3.60	0.20	28.61
0.14°	0.29	21.49	0.04	139.54
	1.75	3.60	0.20	28.27

Supplementary Table 12 | Incident-angle-dependent peak parameters for thermally annealed DPE NP films. (100) and (010) peak position q , d-spacing, FWHM and CCL at different incident angles.

DPE	q ($\text{\AA}^{-1}$)	d -spacing ($\text{\AA}$)	FWHM ($\text{\AA}^{-1}$)	CCL ($\text{\AA}$)
0.11°	0.29	21.55	0.04	131.06
	1.75	3.59	0.19	29.73
0.12°	0.29	21.50	0.04	131.63
	1.75	3.59	0.19	29.64
0.13°	0.29	21.56	0.04	132.66
	1.75	3.59	0.19	29.53
0.14°	0.29	21.49	0.04	133.18
	1.75	3.59	0.19	29.49

Supplementary Table 13 | Photovoltaic performance parameters of PM6:Y6 NP-OSCs annealed at different temperatures under AM 1.5G 100 mW cm⁻² illumination.

Annealing temperature (°C)	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE ^a (%)
No annealing	0.717	13.24	43.06	4.08 (3.69±0.31)
140	0.769	19.07	53.93	7.90 (7.44±0.36)
180	0.758	22.54	60.19	10.28 (9.81±0.38)
200	0.752	22.93	62.14	10.71 (10.34±0.24)
220	0.735	22.41	63.20	10.45 (10.23±0.20)
250	0.592	10.44	51.59	3.18 (2.86±0.23)

^a Values in parentheses represent average parameters and standard deviation obtained from 5 individual devices.

Supplementary Table 14 | Photovoltaic performance parameters of PM6:Y6 NP-OSCs annealed at 200 °C for different times under AM 1.5G 100 mW cm⁻² illumination.

Annealing time (min)	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE ^a (%)
5	0.760	21.96	60.24	10.06 (9.78±0.24)
10	0.752	22.93	62.14	10.71 (10.34±0.24)
15	0.713	23.44	58.75	9.82 (9.45±0.36)

^a Values in parentheses represent average parameters and standard deviation obtained from 5 individual devices.

Supplementary Table 15 | Photovoltaic performance parameters of PM6:Y6 NP-OSCs prepared from inks concentrated from different CF:water volume ratios under AM 1.5G 100 mW cm⁻² illumination.

CF:water (v/v)	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE ^a (%)
1:5	0.749	22.22	56.79	9.45 (8.99±0.45)
1:6	0.752	22.93	62.14	10.71 (10.34±0.24)
1:7	0.768	21.14	60.87	9.89 (9.41±0.33)

^a Values in parentheses represent average parameters and standard deviation obtained from 6 individual devices.

Supplementary Table 16 | Photovoltaic performance parameters of PM6:Y6 NP-OSCs prepared with different 1-PN volume fractions under AM 1.5G 100 mW cm⁻² illumination.

1-PN (v/v %)	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE ^a (%)
1	0.738	22.66	60.52	10.17 (9.86±0.22)
1.5	0.730	22.11	61.12	9.86 (9.37±0.20)
2	0.715	21.65	58.32	9.03 (8.63±0.23)

^a Values in parentheses represent average parameters and standard deviation obtained from 5 individual devices.

Supplementary Table 17 | Photovoltaic performance parameters of PM6:Y6 NP-OSCs prepared with different DPE volume fractions under AM 1.5G 100 mW cm⁻² illumination.

DPE (v/v %)	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE ^a (%)
1	0.773	24.23	64.74	12.18 (11.75±0.34)
1.5	0.767	24.82	66.40	12.64 (12.15±0.24)
2	0.752	24.75	62.44	11.65 (11.26±0.25)

^a Values in parentheses represent average parameters and standard deviation obtained from 6 individual devices.

Supplementary Table 18 | Photovoltaic performance parameters of as-cast PM6:Y6 NP-OSCs processed with and without additives under AM1.5G 100 mW cm⁻² illumination.

As-cast	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE ^a (%)
Control	0.717	13.24	43.06	4.08 (3.69±0.31)
1-PN	0.545	16.01	40.75	3.56 (3.28±0.21)
DPE	0.744	18.84	44.23	6.20 (5.66±0.31)

^a Values in parentheses represent average parameters and standard deviation obtained from 5 individual devices.

Supplementary Table 19 | Literature summary of reported NP-OSCs and corresponding references.

Sample	Synthesis method	Dispersion medium	Solvent	PCE (%)	Year	Ref.
PM6:Y6	Miniemulsion	water	CF	12.64	2026	This work
PM6:Y6	Miniemulsion	water	CF	10.71	2026	This work
PM6:Y6	Miniemulsion	water	CF	9.86	2026	This work
P1:PCBM	Miniemulsion	water	CF	0.55	2011	3
P3HT:PCBM	Miniemulsion	water	CF	0.29	2012	4
PFB:F8BT	Miniemulsion	water	CF	0.39	2012	5
P3HT:PCBM	Miniemulsion	water	CF	1.31	2013	6
P3HT:ICBA	Miniemulsion	water	CF	2.50	2013	7
P3HT:PCBM	Miniemulsion	water	CF	1.20	2014	8
P3HT:PCBM	Miniemulsion	water	CF	2.15	2014	9
P3HT:ICBA	precipitation	MeOH	CF	4.10	2014	10
P3HT:PCBM	Miniemulsion	water	CF	1.16	2015	11
PSBTBT:PCBM	Miniemulsion	water	CF	1.30	2015	12
P(TBT-DPP):ICBA	Miniemulsion	water	CF	2.63	2015	13
TQ1:PC71BM	Miniemulsion	water	CF	2.54	2016	14
P3HT:ICBA/P3HT	precipitation	EtOH	CF	4.20	2016	15
P3HT:ICBA	precipitation	EtOH	CF	4.30	2016	16
PCDTBT:PC ₇₁ BM	Miniemulsion	water	DCB	1.90	2017	17
PDPP5T:PC71BM	Miniemulsion	water	CF	2.38	2017	18

PBDTTPD:PC71 BM	Miniemulsion	water	CB	3.80	2017	19
PTNT:PC71BM	Miniemulsion	water	o-XY	1.65	2018	20
PDPP5T- 2:PC71BM	Miniemulsion	water	CF	3.38	2018	21
P3HT:ICBA	nanoprecipitati on	EtOH	CF	4.52	2018	22
PBQ-QF:ITIC	nanoprecipitati on	water	THF	7.50	2018	23
P3HT:PCBM	Miniemulsion	water	CF	1.20	2019	24
P3HT:ICxA	Miniemulsion	water	CF	1.51	2019	25
PBDTT- FTTE:PC60- PCBM	Miniemulsion	water	CF	5.39	2019	26
P3HT:ICBA	Miniemulsion	water	CF	1.25	2020	27
P3HT:PCBM	Miniemulsion	water	CF	1.78	2021	28
P3HT:ICBA	Miniemulsion	MeOH/AcO H	CF	3.50	2021	29
PTB7- <i>b</i> -P4VP: PC ₇₁ BM	Miniemulsion	Water	TL/ o-XY	0.85	2022	30
P3HT:PCBM	Miniemulsion	Water	CF	1.55	2022	31
PTB7:PC ₇₁ BM	Miniemulsion	Water	CF	2.03	2022	32
P3HT:ICBA	nanoprecipitati on	EtOH	CF	3.68	2022	33
P3HT:ICBA	nanoprecipitati on	EtOH	CF	4.00	2022	34
J71:Y6	nanoprecipitati on	Acetonitrile	CF	10.6	2022	35

PM6:BTP-eC9	nanoprecipitation	Water	THF	11.1	2022	36
PTB7-Th: PC ₇₁ BM	Miniemulsion	Water	CF	5.11	2023	37
PTQ10:Y6	Miniemulsion	Water	CF	9.98	2023	38
P3HT:o-IDTBR	Miniemulsion	Water	CF	1.86	2024	39
P3HT:PCBM	nanoprecipitation	Water SDS/F127	THF	2.44	2024	40
PTQ10:Y6	nanoprecipitation	Water	THF	10.14	2024	41
P3HT:PC ₇₁ BM	nanoprecipitation	EtOH/Water Surfactant-free	CF	2.7	2025	42
PTzBI-dF:L8BO	Miniemulsion	Water/SDS	CF	9.45	2025	43
FO8-T:Y6	Miniemulsion	Water/SDS	CF	10.64	2026	44

Supplementary Table 20 | Photovoltaic performance parameters of PM6:Y6 NP-OSCs fabricated under different relative humidities (RH) under AM 1.5G 100 mW cm⁻² illumination.

RH	V_{oc}	J_{sc}	FF	PCE ^a
(%)	(V)	(mA cm ⁻²)	(%)	(%)
30	0.771	24.87	66.32	12.77 (12.24±0.35)
40	0.767	24.82	66.40	12.64 (12.15±0.24)
50	0.761	24.75	65.92	12.41 (12.10±0.24)
60	0.754	25.04	64.36	12.15 (11.79±0.34)

^a Values in parentheses represent average parameters and standard deviation obtained from 6 individual devices.

Supplementary Table 21 | The FoM θ , FoM α , L_{dri} , and L_{diff} for PM6:Y6 NP-OSCs

	FoM θ	FoM α	L_{dri} (nm)	L_{diff} (nm)
Control	0.0021	0.7942	2598.5	106.8
1-PN	0.0041	0.9103	1885.0	93.2
DPE	0.0019	0.7181	2744.8	118.2

Supplementary Table 22 | Lifetimes and time-integrated fractional contributions obtained by fitting the Y6 GSB kinetic trace at 820 nm for blend films prepared under control, 1-PN and DPE conditions. ^a

Probe @ 820 nm	τ_1 (ps)	τ_2 (ps)	τ_3 (ps)
Control	1.37 ± 0.15 (0.9%)	7.77 ± 0.74 (4.4%)	318.6 ± 26.0 (94.7%)
1-PN	0.79 ± 0.07 (0.5%)	9.52 ± 0.68 (4.0%)	414.3 ± 42.7 (95.5%)
DPE	1.51 ± 0.09 (2.4%)	18.74 ± 2.02 (12.8%)	348.7 ± 82.7 (84.8%)

^a The fitted lifetimes, fitting standard error, and corresponding proportions.

Supplementary Table 23 | Lifetimes and time-integrated fractional contributions obtained by fitting the Y6 LE kinetic trace at 900 nm for blend films prepared under control, 1-PN and DPE conditions. ^a

Probe @ 900 nm	τ_1 (ps)	τ_2 (ps)	τ_3 (ps)
Control	0.26 ± 0.02 (2.9%)	2.71 ± 0.16 (11.6%)	54.72 ± 4.03 (85.5%)
1-PN	0.43 ± 0.02 (2.0%)	6.82 ± 0.44 (9.8%)	168.92 ± 17.86 (88.2%)
DPE	0.17 ± 0.01 (2.7%)	2.06 ± 0.11 (10.0%)	47.80 ± 2.98 (87.3%)

^a The fitted lifetimes, fitting standard error, and corresponding proportions.

Supplementary Table 24 | The detailed energy loss data of PM6:Y6 NP-OSCs prepared under control, 1-PN and DPE conditions.

	E_g	V_{OC}	E_{loss}	V_{OC}^{SQ}	ΔE_1	V_{OC}^{rad}	ΔE_2	ΔE_3
	(eV)	(V)	(eV)	(V)	(eV)	(V)	(eV)	(eV)
Control	1.392	0.752	0.640	1.12	0.272	1.042	0.078	0.290
1-PN	1.397	0.730	0.667	1.14	0.257	1.045	0.095	0.315
DPE	1.391	0.767	0.624	1.12	0.271	1.034	0.086	0.267

Supplementary References

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