

Supporting information

Supramolecular engineering in hybrid perovskite optoelectronics

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Table S1. Summary of the photovoltaic parameters for the reported supramolecular engineering PSCs

Supramolecular Agent	Device Structure	Enhancement ratio (%)	Control PCE (%)	PCE (%)	Voc (V)	Jsc (mA/cm ²)	FF (%)	Stability	Year
Phenylalkylamine	n-i-p PSC (FTO/TiO ₂ /PVSK/Spiro-OMeTAD/Au)	10.98	17.3	19.2	1.12	23.6	73.0	Retained 50% of PCE after more than four months' exposure to moisture air	2016 ¹
Mercapto-tetrazolium (S) and phenylammonium (N) moieties	n-i-p PSC (FTO/cp-TiO ₂ /mp-TiO ₂ /PVSK/Spiro-OMeTAD/Au)	8.85	19.2	20.9	1.15	24.0	75.0	Retained 98.1% of PCE after 1000 h under full solar light soaking at 60 °C in Ar (unencapsulated)	2018 ²
5-ammonium valeric acid iodide (AVAI)	n-i-p PSC (FTO/TiO ₂ /FAPbI ₃ /Spiro-OMeTAD/Au)	160.88	7.3	18.9	1.08	25.1	70.0	Retained 90% of PCE after 300 h under MPP tracking with white light illumination	2019 ³
Dibenzo 24-crown-8 (DB24C8)	n-i-p PSC (FTO/cp-TiO ₂ /mp-TiO ₂ /PVSK/Spiro-OMeTAD/Au)	10.23	21.5	23.7	1.15	25.8	79.5	Retained >80% of PCE after 300 h under MPP tracking with full solar illumination (AM 1.5 G, 100 mW/cm ² in N ₂ , 25°C).	2020 ⁴

								(unencapsulated)	
2D black phosphorene (BP)	n-i-p PSC (FTO/TiO ₂ /PVSK/Spiro-OMeTAD/Ag)	16.99	17.0	19.8	1.12	23.9	73.8	-	2020 ⁵
Chloropropyltrimethoxysilane (CPS)	n-i-p PSC (FTO/PEDOT:PS S/PVSK/Spiro-OMeTAD/Ag)	18.71	17.1	20.3	1.12	24.8	73.0	Retained ~85% PCE after 30 days at RH=35%	2021 ⁶
18-crown-6 (18C6)	n-i-p PSC (FTO/ZnO-ZnS/TiO ₂ /PVSK/Spiro-OMeTAD/Au)	20.12	17.1	20.5	1.06	24.4	79.2	Retained 92% of PCE after 1000 h under ambient condition	2021 ⁷
NiP-supramolecule	n-i-p PSC (FTO/modified ZnO/mp-TiO ₂ /PVSK/phthalocyanine/Au)	5.78	22.8	24.2	1.17	25.5	81.01	Retained 90% of PCE after 5000 h at RH=65% and room temperature; Retained 90% of PCE after 3000 h at 85 °C with RH~55%	2021 ⁸
2-phenylethylammonium (PEA ⁺) and 2(perfluorophenyl)ethylammonium (FEA ⁺) moieties	n-i-p PSC (FTO/cp-TiO ₂ /mp-TiO ₂ /PVSK/Spiro-OMeTAD/Au)	10.20	19.6	21.6	-	-	-	Retained ~90% of PCE after 100 h of operation	2021 ⁹
ortho-isomers of (phenylene)di(ethylammonium) iodide (o-PDEAI ₂)	n-i-p PSC (FTO/cp-TiO ₂ /mp-TiO ₂ /PVSK/Spiro-OMeTAD/Au)	9.02	21.9	23.9	1.16	24.8	83.5	Retained 85% of PCE under an RH=40–50% (unencapsulated); Retained 75% of PCE after 1000 h at 85 °C in a nitrogen atmosphere (unencapsulated)	2021 ¹⁰
15-crown-5 (15C5)	n-i-p PSC (FTO/cp-TiO ₂ /PVSK/Spiro-OMeTAD/Au)	12.51	19.2	21.6	1.16	24.2	76.6	Retained >90% of PCE after 1000 h in the dark under RH=35–40% (unencapsulated)	2021 ¹¹
Dibenzo-21-crown-7 (DB21C7)	n-i-p PSC (FTO/cp-TiO ₂ /mp-TiO ₂ /PVSK/Spiro-OMeTAD/Au)	14.62	21.2	24.3	1.17	25.5	81.9	Target perovskite film was stable in air for 380 days, while the control film degraded completely within 5 days	2021 ¹²
3D star-shaped polyhedral oligomeric silsesquioxane-poly(trifluoroethyl methacrylate)-b-poly(methyl methacrylate) (PPP) polymer	p-i-n PSC (ITO/NiO _x /PVSK+PPP/PCBM+C60/BCP/Cr/Au)	18.74	18.6	22.1	1.13	23.2	84.1	Retained ~93% of PCE after 6000 h under a RH= 40% (unencapsulated)	2021 ¹³
β-diketone based ligand, N,N,N',N'-tetraphenylmalondiamide [TPMA]	p-i-n PSC (ITO/PEDOT:PSS/Pb-Sn PVSK/C60/BCP/Ag)	10.94	18.6	20.7	0.82	33.0	76.2	Retained 94% of PCE after 1000 h in N ₂ glovebox and only 8% degradation after continuously heated for 100 h at 80 °C (unencapsulated)	2021 ¹⁴
Carbazole-based material	n-i-p PSC (FTO/TiO ₂ /PVSK/	-1.03	19.5	19.3	1.18	21.7	75.0	Retained >90% of PCE after 550 h	2021 ¹⁵

(C ₆₂ H ₅ F ₄ IN ₃ O ₃) (PFI)	HTMs/Au)							under MPP tracking with a TS80 of ≈2600 h	
1-butyl-3-methylimidazolium-based ILs ([BMIM]X)	n-i-p PSC (FTO/SnO ₂ /(FAPbI ₃) _{0.95} (MAPbBr ₃) _{0.05} /Spiro-OMeTAD/Au)	6.65	21.9	23.4	1.20	24.7	79.0	Retained 95% of PCE after 4080 h in ambient dry-air storage and 80% of PCE after 1400 h continuous light illumination	2022 ¹⁶
Chenodeoxycholic acid (CDCA)	n-i-p PSC (ITO/SnO ₂ /PVSK/CDCA/Spiro-OMeTAD/Au)	11.51	20.9	23.3	1.16	25.4	78.9	Retained 92% of PCE after 1600h under ambient conditions (unencapsulated); Almost unchanged after heating at 85 °C for 500 h in a nitrogen atmosphere (unencapsulated)	2022 ¹⁷
2, 4, 6-tris(4-aminophenyl)-s-triazine (TAPT)	p-i-n PSC (ITO/NiO _x /PTAA/TAPT/PVSK/PCBM+C60/BCP/Cr/Au)	9.88	22.4	24.6	1.16	26.1	81.2	Retained 89.7% of PCE after 1500 h of maximum power point tracking and 91.9 % of PCE after 1065 h of 85 °C heat treatment	2023 ¹⁸
Potassium cation-18-crown ether-6 complexes (18C6-K ⁺)	n-i-p PSC (FTO/SnO ₂ +18C6 + K ₂ SO ₄ /PVSK/Spiro-OMeTAD/Ag)	6.52	20.3	21.6	1.13	24.1	79.5	Retained ~90% of PCE after 500h storage in dry air	2023 ¹⁹
1-aza-18-crown-6 (A18C6)	n-i-p PSC (FTO/SnO ₂ /PVSK / A18C6/Spiro-OMeTAD/ Au)	9.91	21.9	24.1	1.17	25.0	82.2	Retained a long T80 lifetime of 510 h under MPP tracking conditions under 1-sun equivalent illumination at around 20 °C in an inert atmosphere (unencapsulated)	2023 ²⁰
3-fluoro-4-methoxy 4',4''-bis((4-vinyl benzyl ether) methyl) triphenylamine (FTPA)	n-i-p PSC (FTO/SnO ₂ FA _{0.95} MA _{0.05} Pb _{(0.95} Br _{0.05}) ₃ / Spiro-OMeTAD/ Au)	7.21	22.5	24.1	1.18	24.4	83.5	Retained >95% of PCE after 1000 h under MPP tracking; Retained 95% of PCE after 2000 h at air ambient of RH~50%	2023 ²¹
biphenyl-4,4'-dithiol (P2)	n-i-p PSC (FTO/cp-TiO ₂ /Li-doped mp-TiO ₂ /PVSK/spiro-OMeTAD/ Au)	5.73	22.7	24.0	1.20	25.4	79.0	Retained ~88% of PCE after 2000 h under MPP tracking at 40 °C and 1 sun illumination	2023 ²²
1,4-diazabicyclo[2.2.2]octane (DABCO)	n-i-p PSC (ITO/ cp-SnO ₂ or DABCO-SnO ₂ /PVSK/ spiro-OMeTAD/ Ag)	15.12	20.5	23.6	1.17	24.8	81.6	Retained 87% of PCE over 1500 h under ISOS-D1 standard conditions	2023 ²³
Ammonia-docked (DCAP)	n-i-p PSC (FTO/NiO _x /Perovskite/PCBM/Ag)	12.37	22.5	25.3	1.15	25.6	85.6	Retained 81.5% of PCE after 1200h under light (1 Sun illumination) and heat (65 °C)	2024 ²⁴
15-crown-5 (15C5)	n-i-p PSC (ITO/PEDOT:PSS	12.10	19.2	21.5	1.16	24.2	76.6	Retained >90% of PCE after 1000 h in	2024 ¹¹

	/PVSK/P3HT/Ag)							the dark under RH=35%–40%; 17% PCE loss at 85°C for 200 h	
Dibenzo-18-crown-6 (DB18C6)	p-i-n PSC (FTO/NiO _x /2PACz/ PVSK/Crown ethers/ PCBM/BCP/Ag)	11.91	20.8	23.3	1.13	24.6	83.5	Retained 92% of PCE after 1224 h of aging in an N ₂ environment (unencapsulated) Retained 89% PCE after 1000 h of aging in RH=30%–40% (unencapsulated)	2024 ²⁵
β cyclodextrin (β-CD)	p-i-n PSC (ITO/SAM(Meo-2PACz)/ PVSK/C60/ BCP/Ag)	10.31	19.4	21.4	1.14	23.5	80.2	Retained >73% of PCE after 320 hours of testing at 50–60 °C. (0.5% of β-CD treated condition)	2024 ²⁶
dibenzo-21-crown-[7] (DB21C7)	n-i-p PSC (FTO/TiO ₂ /PVSK/ Crown ethers/Spiro-OMeTAD/Au)	18.00	5.0	5.9	1.5	5.5	73.0	Retained ~80% of PCE over 300 h at 85 °C under nitrogen atmosphere, or ambient temperature	2024 ²⁷
Benzo-18-crown-6-ether (B18C6)	n-i-p PSC (ITO/SnO ₂ /PVSK/ PEABr/spiro-OMeTAD/ Au)	5.64	19.5	20.6	1.21	20.5	83.0	Retained 99.6% of PCE after 1080 hours of storage in air (unencapsulated)	2024 ²⁸
Alkylthiophene-substituted polythiophenes (PT4T-2F)	n-i-p PSC (FTO/SnO ₂ /FA _{0.83} Cs _{0.17} Pb(I _{0.7} Br _{0.3}) ₃ / Interlayer/Spiro-OMeTAD/Au)	8.11	17.8	19.2	1.23	20.9	75.1	Retained 83% of PCE over 26 days under RH=45-50%	2024 ²⁹
1-aza-15-crown5-ether (A15C5)	n-i-p PSC (FTO/SnO ₂ /PVSK /A15C5/Spiro-OMeTAD/Ag)	10.84	21.8	24.1	1.14	25.3	83.6	Retained ~96.5% of PCE after 960 h in ambient air with RH=40 ± 10% (unencapsulated) Retained 75.3% of PCE after 360 h under N ₂ atmosphere and 85 °C (unencapsulated)	2024 ³⁰
15-crown-5 + CDT-S and CDT-N,	n-i-p PSC (FTO/cp-TiO ₂ /mp-TiO ₂ /PVSK/Spiro-OMeTAD/Au)	8.88	21.2	23.1	1.15	24.5	81.6	Retained 90.5% of PCE after 1000 hours of storage in the dark (unencapsulated); Retained 80.83% under MPP tracking for 1000 h (unencapsulated)	2024 ³¹
Crown ether derivative (PC15)	n-i-p PSC (ITO/SnO ₂ /PVSK/ PC15/Spiro-OMeTAD/Ag)	19.23	20.8	24.8	1.19	25.5	81.6	Retained 86% of PCE after 1500 h under MPP tracking with continuous illumination (1-sun) and a temperature of 25 ± 5 °C	2024 ³²
PEAI+DB21C7	n-i-p PSC (FTO/cp-TiO ₂ /mp-TiO ₂ /PVSK/Spiro-OMeTAD/Au)	12.54	22.6	25.4	1.18	25.8	83.4	Retained > 96.6% of PCE after 1050 h of continuous operation under 1 sun illumination	2024 ³³
benzo[c][1,2,5]thiadiazol-4-	n-i-p PSC (FTO/cp-TiO ₂ /mp-	5.91	18.6	19.7	-	-	-	Retained >80% of PCE after 1000h	2024 ³⁴

methylammonium (BTDZ) halide (X = I, Br)	TiO ₂ /Cs _{0.05} FA _{0.90} MA _{0.05} Pb(I _{0.95} Br _{0.05}) ₃ / BTDZ/Spiro-OMeTAD/Au)							under MPP tracking	
Benzo-18-crown-6 (B18C6)	n-i-p PSC (FTO/ cp-TiO ₂ /mp-TiO ₂ /PVSK/spiro-OMeTAD/ Ag)	11.30	19.3	21.5	1.17	24.8	74.0	Retained 80% of PCE after 300 h at RH =85%	2024 ³⁵
P,M-(1-methylene-3-methyl-imidazolium)[6]helicene iodides	n-i-p PSC (FTO/ cp-TiO ₂ /mp-TiO ₂ /PVSK/spiro-OMeTAD/ Ag)	-0.48	20.9*	20.8*	1.10*	24.1*	76.8*	Retained 80% of PCE after 1000 h under MPP tracking with full solar illumination (AM 1.5 G, 100mWcm ⁻² , N ₂ , and 25°C). (unencapsulated)	2024 ³⁶
2,2,3,3,3-pentafluoropropylamine hydrochloride (PFPAcl) and 3,3,3-trifluoropropylamine hydrochloride (TFPAcl)	n-i-p PSC (ITO/SnO ₂ /PVSK/spiro-OMeTAD/Au)	13.20	20.8	23.6	1.17	24.7	81.5	Retained 89.8% of PCE after 1000 h under MPP tracking with 100 mWcm ⁻² light illumination provided by white LED and in N ₂ (unencapsulated)	2024 ³⁷
18-crown-6 ether (18C6)	n-i-p PSC (FTO/TiO ₂ /PVSK/Spiro-OMeTAD/Au)	8.74	20.4	22.1	1.24	21.3	84.1	Retained 95% of PCE after 1500 h under MPP tracking	2025 ³⁸
Phenylethylammonium acetate (PEAAc)	n-i-p PSC (FTO/SnO ₂ /PVSK / passivation layer/ Spiro-OMeTAD/Au)	10.18	22.6	24.9	1.19	26.5	78.7	Retained 86% of PCE after 500 h at 85 °C in a glove box without illumination (unencapsulated)	2025 ³⁹

Note: *average data; Enhancement ratio (%)= (PCE_{treated} – PCE_{control}) / PCE_{control} *100%

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