Supporting Information

N-type Polymeric Mixed Conductors for All-in-One Aqueous Electrolyte Gated Photoelectrochemical Transistors

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Figure S1. CV curves of a) $p(C_6-NDI-T)$, b) P-90, c) BBL, d) gAIID-2FT, and e) P-75 films recorded in degassed acetonitrile, 0.1 M TBA-NPF₆, with a scan rate of 50 mV/s. CV curve in f) was recorded with 0.01 M ferrocene and 0.1 M TBA-NPF₆ in degassed acetonitrile. Multiple cycles are shown (overlapping), suggesting the chemical stability of the polymer films in this electrolyte and measurement range.

Table S1. LUMO level determination using the reduction onset extracted from the CV curves in Figure S1.

	Eonset, red (V vs Ag/AgCl)	E _{LUMO} (eV)
$p(C_6NDI-T)$	- 0.207	- 4.405
P-90	- 0.377	- 4.235
BBL	- 0.220	- 4.392
gADII-2FT	- 0.421	- 4.191
P-75	+0.244	- 4.856



Figure S2. The evolution of the UV-Vis spectra of **a**) p(C6NDI-T), **b**) P-90, **c**) BBL, **d**) gAIID-2FT, and **e**) P-75 films as they were cycled from 0.2 V to -0.7 V vs. Ag/AgCl in PBS. Each spectrum was collected 2 minutes after the doping bias was applied with a 0.1 V increment. The polymer films had similar thicknesses of around 100 nm.



Figure S3. CV curves of the films recorded in air and N_2 . The scan rate was 50 mV/s. The arrow in a) indicates the direction of the scans. The inset in d) zooms into the voltammogram recorded in N_2 .



Figure S4. a) The OCP-time profile of P-75 in PBS during the application of two light pulses (10 seconds ON and 50 seconds OFF, with a power of 0.258 W/cm²). **b)** The OCP time profile of $p(C_6NDI-T)$ when shining the light from the polymer or the ITO side. **c)** The current-time profile of the films measured at 0 V vs. OCP in dark and light conditions. The light was ON for 0.5 seconds and OFF for 10 seconds.



Figure S5. Nyquist plots of a) $p(C_6NDI-T)$, b) P-90, c) BBL, and d) P-75 films recorded at 0 V vs. OCP in dark and light conditions.

Table S2. The light intensity values correspond to the symbols used in Figures 2, 4, and 5.

Symbol	Corresponding Light Intensity (W/cm ²)
I ₁	0.003
I_2	0.007
I_3	0.019
I_4	0.039
I_5	0.078
I ₆	0.154
I_7	0.258



Figure S6. TA spectroscopy of dry $p(C_6NDI-T)$ films on bare glass and ITO/glass (data originally published in ¹). **a)** TA spectra of $p(C_6NDI-T)/ITO$ at different pump-probe delay times at a fluence of 24 μ J/cm². **b)** TA spectra of intrinsic $p(C_6NDI-T)$ film on glass (red) and $p(C_6NDI-T)/ITO$ (blue) recorded at different fluences and averaged over pump-probe delay times of 0-1 ps. The pump wavelength was 660 nm. **c)** picosecond-nanosecond kinetics of the $p(C_6NDI-T)/ITO$, averaged over different spectral ranges. Overlaid are the multi-exponential decay fits (see Table S3).



Figure S7. TA spectroscopy of dry P-90 films on bare glass and ITO/glass. **a)** TA spectra of P-90/ITO at different pump-probe delay times at a fluence of 18 μ J/cm². **b)** TA spectra of intrinsic P-90 films on glass (red) and P90/ITO (blue) at different fluences, averaged over pump-probe delay times of 0-1 ps. The pump wavelength was 660 nm. **c)** picosecond-nanosecond kinetics of the P-90/ITO, averaged over different spectral ranges. Overlaid are the multi-exponential decay fits (see **Table S3**).



Figure S8. TA spectroscopy of dry BBL films on bare glass and ITO/glass. **a)** TA spectra of BBL/ITO at different pump-probe delay times at a fluence of $18 \,\mu$ J/cm². **b)** TA spectra of intrinsic BBL films on glass (red) and BBL/ITO (blue) at different fluences, averaged over pump-probe delay times of 0-1 ps. The pump wavelength was 660 nm. **c)** picosecond-nanosecond kinetics of the BBL/ITO b, averaged over different spectral ranges. Overlaid are the multi-exponential decay fits (see **Table S3**).



Figure S9. TA spectroscopy of dry gAIID-2FT films on bare glass and on ITO/glass. **a)** TA spectra of gAIID-2FT/ITO at different pump-probe delay times at a fluence of $18 \mu J/cm^2$. **b)** TA spectra of intrinsic gAIID-2FT films on glass (red) and gAIID-2FT/ITO (blue) at different fluences, averaged over pump-probe delay times of 0-1 ps. The pump wavelength was 660 nm. **c)** picosecond-nanosecond kinetics of the gAIID-2FT/ITO, averaged over different spectral ranges. Overlaid are the multi-exponential decay fits (see **Table S3**).



Figure S10. TA spectroscopy of dry P-75 films on bare glass and ITO/glass. **a)** TA spectra of P-75/ITO at different pump-probe delay times at a fluence of 18 μ J/cm². **b)** TA spectra of intrinsic P-75 films on glass (red) and P-75/ITO (blue) at different fluences, averaged over pump-probe delay times of 0-1 ps. The pump wavelength was 660 nm. **c)** picosecond-nanosecond kinetics of the P-75/ITO, averaged over different spectral ranges. Overlaid are the multi-exponential decay fits (see **Table S3**).



Figure S11. Doping-induced absorption (DIA) spectra of the n-type films.

Table S3. Decay parameters obtained from fitting the picosecond-nanosecond TA kinetics (**Figures 3b and S5-S10**) for different species to the equation: $\Delta T/T = A_1 e^{(-t/\tau_1)} + A_2 e^{(-t/\tau_2)}$ where *t* is the pump-probe delay time, A_i are the amplitudes of the decay components, t_i are the lifetimes of the decay components, and t_{ave} is the amplitude-averaged lifetime. Photobleach (PB) kinetic fits are included for gAIID-2FT and P-75.

Material	species	wavelength	y ₀	A_0	τ_1	A ₂	τ_2	τ_{ave}
		(nm)			(ps)		(ps)	(ps)
$P(C_6NDI-T)$	charge		0.0385	0.2058	18.28	0.6826	170.53	135.26
	exciton		0.0000	0.3801	7.79	0.5790	134.43	84.24
	ICT		0.0000	0.1626	4.38	0.7518	246.30	203.29
P90	charge	850-940	-0.0771	0.2181	0.75	0.9038	45.13	36.50
	exciton	1020-1150	0.0385	0.2347	2.52	0.6603	41.26	31.10
	ICT	1300-1500	0.1086	0.1598	2.83	0.6930	40.44	33.39
BBL	charge	720-780	0.2187	0.5292	18.42	0.2655	243.11	93.48
	exciton	1030-1230	0.0593	0.6389	1.02	0.3628	11.78	4.92
	ICT	1250-1400	0.1340	0.4814	2.79	0.3073	39.34	17.03
gAIID-2FT	PB	680-730	0.0788	0.3575	7.76	0.5224	133.60	82.47
	charge	800-890	0.1608	0.8508	144.00	0.8508	144.00	144.00
	exciton	1050-1350	0.0034	0.3814	11.05	0.5722	185.34	115.63
P-75	PB_1	630-660	0.0371	1.1971	0.31	0.1155	1.96	0.45
	PB ₂	950-1050	0.0330	1.2587	0.31	0.1251	2.14	0.48
	PA ₂	1300-1550	0.0336	1.5035	0.21	0.0899	1.67	0.30



Figure S12. THz spectroscopy of dry P-75 film on a quartz substrate. **a)** THz-time-domain spectroscopy (THz-TDS) of P-75 film and quartz substrate. **b)** Fast Fourier Transform (FFT) of THz-TDS waveforms. **c)** Real and imaginary parts of the complex conductivity of the P-75 film, with an average value of 190.8 Ω cm⁻¹. **d)** Optical pump-THz probe kinetics after photoexcitation with 500 nm pump (1.26 mJ/cm²). Overlaid in red is a mono-exponential decay fit to the data, giving a lifetime of 935 fs. **e)** TDS of the P-75 film before (black) and after (red) photoexcitation. Note the photoexcited TDS waveform is scaled by a factor of 50. **f)** FFT of the ground and photoexcited THz-TDS waveforms. **g)** Real and imaginary parts of the complex photoconductivity of the P-75 film (average value = 2.26Ω cm⁻¹). **h)** Mobility of P-75 from the THz measurements (average value = 9.0×10^{-3} cm² V⁻¹ s⁻¹).



Figure S13. **a)** The transfer curves, i.e., the drain current (I_{DS}) vs. gate voltage (V_{GS}), of OECTs collected in the dark and light (660 nm, 0.258 W/cm²) conditions, in forward and backward scan. **b)** The threshold values (V_{th}) in dark and light conditions, as determined from the square root of I_{DS} vs. V_{GS} . **c)** The transfer curves were obtained for channels gated with an Ag/AgCl gate electrode. The data were collected under both dark and light conditions (660 nm, 0.258 W/cm²), in forward and backward scan.



Figure S14. a) The transfer curves, b) the transconductance vs. V_{GS} , and c) the square root of I_{DS} vs. V_{GS} plots of P-75 collected in the dark and under light.



Figure S15. The real-time change in I_{GS} and I_{DS} for **a**) $p(C_6NDI-T)$, **b**) P-90, **c**) BBL, and **d**) gAIID-2FT OECTs upon light pulses at 0.258 W/cm² measured in O₂-depleted environment. The light was ON for 1 minute and OFF until the current returned to its value in the dark state.



Figure S16. The channel current response to a square-shaped voltage pulse at the gate electrode (0.4 V is applied until the current is stabilized at a constant V_{DS} =0.4 V). The devices have a) p(C₆NDI-T), **b**) P-90, **c**) BBL and **d**) gAIID-2FT as a channel gated with Ag/AgCl. The devices were evaluated under dark.



Figure S17. The channel current response to a square-shaped voltage pulse at the gate electrode (0.4 V is applied until the current is stabilized, at a constant $V_{DS}=0.4$ V) for **a**) p(C₆NDI-T), **b**) P-90, **c**) BBL and **d**) gAIID-2FT OECTs. The devices were evaluated under dark.

Polymer	Ag/AgCl gate (ms)	Polymer gate (ms)	
p(C ₆ NDI-T)	3.6	38.9	
P-90	6.9	26.9	
BBL	1.5	220.2	
gAIID-2FT	6.7	18.0	

Table S4. A comparison of the OECT response (switching on) time when the gate is an Ag/AgCl or the polymer film on Au.

Polymer	I _{DS} ON (s)	I _{DS} OFF (s)
$p(C_6NDI-T)$	4.27	18.4
P-90	5.02	13.54
BBL	2.40	42.7
gAIID-2FT	4.50	3.50

Table S5. The light induced rise and fall times obtained from Figures 18-21 (extracted from 30s pulses).



Figure S18. The effect of different light pulse lengths on the current change of $p(C_6NDI-T)$ OECTs, when the light at 0.258 W/cm² was ON for **a**) 5 s, **b**) 10 s, **c**) 30 s, and **d**) 60 s.



Figure S19. The effect of different light pulse lengths on the current change of P-90 OECTs, when the light at 0.258 W/cm² was ON for **a**) 5 s, **b**) 10 s, **c**) 30 s, and **d**) 60 s.



Figure S20. The effect of different light pulse lengths on the current change of BBL OECTs, when the light was at 0.258 W/cm^2 was ON for **a**) 5 s, **b**) 10 s, **c**) 30 s, and **d**) 60 s.



Figure S21. The effect of different light pulse lengths on the current change of gAIID-2FT OECTs, when the light was at 0.258 W/cm^2 was ON for **a**) 5 s, **b**) 10 s, **c**) 30 s, and **d**) 60 s.



Figure S22. Detectivity of n-type OPECTs as a function of VGS under deep red light with 0.258 W/cm^2 . $V_{DS} = 0.4 V$.

Publication		Materials, mechanism and device performance								
Reference (Year)	Wavelength	Power	Photo-active material	Channel material	Medium	Photo- conversion mechanism	Device operation	I _{DS} (light)/ I _{DS} (dark)	g _m (light)/ g _m (dark)	
² (2022)	Xe lamp	-	PEDOT-Fe- MOF/ ITO gate	PEDOT:PSS	PBS	Faradaic photoinduced electron transfer at the p-n interface then collected at the ITO gate	Depletion	2.0	-	
³ (2023)	425 nm	40 mW.m ⁻²	photosensitive MOF + TiO ₂ nanorods/ FTO gate	PEDOT:PSS	PBS	Faradaic Photoinduced electrons transferred from the conduction band of MOF to TiO ₂ NRs and then to the FTO gate	Depletion	2.4	1.12	
4 (2018)	White light	2000 mW.m ⁻²	Polythiophene / gold gate	PEDOT:PSS	0.1 M NaCl + O ₂ required	Faradaic Light enhanced- oxygen reduction reaction	Depletion	1.6	-	
⁵ (2018)	420 nm	0.2 mW.cm ⁻²	Cds QDs /ITO gate	PEDOT:PSS	PBS + 0.1M ascorbic acid	Faradaic Photoexcited electrons are transferred from the conduction band of Cds QDs to the ITO gate. Electron donor (ascorbic acid) compensates for the excited valance band of Cds	Depletion	6.0	-	
⁶ (2022)	425 nm	5 W lamp	Cds QDs /ITO gate	PEDOT:PSS	PBS + 0.05M ascorbic acid	Faradaic Photoexcited electrons are transferred from the conduction band of Cds QDs to the ITO gate. Electron donor (ascorbic acid) compensates for the excited valance band of Cds	Depletion	2.0	-	

 Table S6. Characteristics and operating conditions of aqueous electrolyte compatible OPECTs.

7 (2022)	White light	0.48 mW.cm ⁻²	CdsIn ₂ S ₄ /FTO gate	PEDOT:PSS	PBS + 0.01M triethanolamine	Faradaic Photoexcited electrons are transferred from the conduction band of CdsIn ₂ S ₄ to the FTO gate. Electron donor (triethanolamine) compensates for the excited valance band	Depletion	2.0	-
⁸ (2022)	White light	5 W lamp	TiO ₂ nanotubes /Ti gate	PEDOT:PSS	Tris-HCl ascorbic acid	FaradaicPhotoexcitedelectron fromdonating ligand (AA) toconduction bandof TIO2 thentransferred to Tigate	Depletion	1.0	-
⁹ (2022)	425 nm	16 mW. cm ⁻²	CdS QDs and TiO ₂ / FTO gate	PEDOT:PSS	1x PBS + ascorbic acid for photo anode	Faradaic Charge separation in CdS QDs, photo-excited electrons are transferred to conduction band of TiO ₂ and then collected in FTO electrode	Depletion	6.8	-
¹⁰ (2022)	425 nm	20 mW. cm ⁻ 2	CdS QDs and TiO ₂ nanotubes / Ti gate	PEDOT:PSS	PBS + electron donor (MEA)	Faradaic Photoexcited electrons are transferred from the conduction band of Cds QDs to TiO ₂ and then transferred to the Ti gate. Electron donor species compensate for the excited valance band of Cds	Depletion	1.4	-
11 (2023)	455 nm	5 W lamp	ZnIn ₂ S ₄ photocatalyst heterojunction	PEDOT:PSS	PBS + 0.05 M ascorbic acid	Faradaic Photoexcited electrons are transferred from the conduction band of ZnIn ₂ S ₄ to the FTO gate. Ascorbic acid compensates for the excited valance band.	Depletion	1.9	-

12 (2023)	455 nm	-	Polymer dot (PDots) on poly(amine) / ITO gate	Dedoped PEDOT:PSS (by amine)	1x PBS + H ⁺ through pH change	Faradaic Photoinduced holes are transferred from the valance band of PDots to the amine HOMO and then to the ITO gate. Photoinduced electrons reduce protons in solution.	Enhancement	2.0	-
¹³ (2023)	365 and 530 nm	365 nm at 3W and 530 nm at 5 mW	OEG-Spiropyran blend at channel / Pt grid as gate	Blend of pgBTTT with the OEG-SP	0.5 M NaCl	Photoisomerizati on of spiropyran compound modulating the channel doping state	Depletion	1.3	-
¹⁴ (2022)	425 nm	-	Cds QDs /ITO gate	PEDOT:PSS	PBS + ascorbic acid	Faradaic Photoexcited electrons are transferred from the conduction band of Cds QDs to the ITO gate. Electron donor compensates for the excited valance band of Cds	Depletion	1.1	-
¹⁵ (2022)	455 nm	-	Cd _x Zn _{1-x} S electrode (metal organic framework)	PEDOT:PSS	PBS + (0.05 M) triethanolamine	Faradaic Photoexcited electrons are transferred from the conduction band of Cd _x Zn ₁ . _x S. Electron donor (triethanolamine) compensates for the excited valance band	Depletion	1.5	-
¹⁶ (2023)	425 nm	-	MXene (Ti ₃ C ₂)/metal organic framework (PCN-224)/NiO/ FTO gate	PEDOT:PSS	1,4- bezoquinone added to electrolyte	$\begin{tabular}{lllllllllllllllllllllllllllllllllll$	Enhancement	1.8	-

17 (2023)	Xe lamp	-	Zn _{1−x} O nanorod arrays/ FTO	PEDOT:PSS	PBS	Faradaic $Zn_{1-x}O$ excited togeneratephotoinducedelectrons thatmoved from thevalance band toconduction band,and then to theFTO	Depletion	3.09 at 0.6 V of V _{GS}	-
¹⁸ (2023)	White light	-	BiVO4/WO3/FTO	PEDOT:PSS	PBS + 50 mM ascorbic acid	Faradaic Photoexcited electrons are transferred between the conduction bands of BiVO ₄ and WO ₃ to FTO electrode. Ascorbic acid neutralize the photogenerated holes and assist the electron transfer	Depletion	1.2	-
¹⁹ (2023)	455 nm	-	pDEB/NiO/FTO	PEDOT:PSS	PBS, PBS + sodium thiophosphat, PBS + alkaline phosphatase or PBS + sodium thiophosphate and alkaline phosphatase	Faradaic Photoexcited holes are transferred pDEB to NiO and then to FTO gate. Simultaneously, photoinduced electrons transferred from conduction band of pDEB to the dissolved O ₂ in the electrolyte	Enhancement	1.9	-
²⁰ (2023)	White LED	3 W	CdTe– BiOBr/ITO	PEDOT:PSS	PBS + 0.1 M ascorbic acid	Faradaic Electrons of BiOBr and CdTe were excited from their valance band to conduction band	Depletion	3.0	-
²¹ (2023)	425 nm	-	Au NPs/(3- mercaptopropyl)t rimethoxysilane (MPTMS)/FTO	PEDOT: PSS	PBS + triethanolamine	Faradaic Ru(II) ground state undergoes excitation to the Ru(II)* state, transferring electrons to a metal contact while transitioning to Ru(III). In the presence of triethanolamine, Ru(III) is then reduced back to Ru(II)	Depletion	1.1	-

22 (2023)	425 nm	-	[Ru(bpy)2(dppz)] 2+/NiO/FTO	PEDOT: PSS	PBS	Faradaic Photoexcited holes are transferred from HOMO of [Ru(bpy) ₂ (dppz)] ² ⁺ to valance band of NiO and then to FTO substrate. Simultaneously electrons are transformed from LUMO of [Ru(bpy) ₂ (dppz)] ² ⁺ to the dissolved oxygen in electrolyte	Enhancement	4.0	-
²³ (2023)	White light	-	Biocatalytic precipitation/ Bi ₂ S ₃ /FTO	PEDOT: PSS	PBS + 0.05 M ascorbic acid	Faradaic	Depletion	1.3	_
24 (2023)	Xe lamp	-	ZnO NEAs/FTO	PEDOT: PSS	PBS	Faradaic Photoexcited electrons are transferred from the conduction band of CdsIn ₂ S ₄ to the FTO gate.	Depletion	3.0	-
²⁵ (2023)	365 nm	0.56 W/cm ² 1.7 W/cm ² and 2.8 W/cm ²	PEDOT:PSS + azobenzenes moieties	PEDOT: PSS	PBS	Integration of photo- switchable molecules modulating the channel de- doping state	Depletion	1.07	-
1 (2023)	660 nm	406 mW.cm ⁻²	p(C ₆ NDI-T)/ gold gate	p(C ₆ NDI-T)	PBS	Potentiometric photovoltage generation at the mixed conductor based gate	Enhancement	428	37.75
²⁶ (2024)	450 nm	28 mW.cm ⁻²	hydrogen-bonded organic frameworks/ TiO ₂ nanoparticles/ FTO gate	PEDOT:PSS	PBS + 100 mM triethanolamine	Faradaic photogenerated electrons of gate were transferred from HOMO to LUMO, then moved to the CB of the TiO2 nanoparticles	Depletion	-	78

27 (2024)	-	-	MXene (Ti ₃ C ₂)/TiO ₂ /ITO	PEDOT: PSS	PBS	Faradaic photogenerated electrons are excited from VB to CB. Ti ₃ C ₂ transfers photogenerated electrons from the TiO ₂ CB to the ITO electrode	Depletion	-	1.5
²⁸ (2024)	Visible light	-	ZnSe/Bi ₂ MoO ₆ nanocomposite	PEDOT: PSS	Tris-HCl + 0.1 M hydroquinone	Faradaic Due to matched energy bands between ZnSe and Bi ₂ MoO ₆ , typical charge transfer process occurred	Depletion	-	1.5
²⁹ (2024)	Visible light	-	MXene/Bi ₂ S ₃ /CdI n ₂ S ₄	PEDOT: PSS	0.1 M Tris– HCl solution	Faradaic typical charge transfer process occurred	Depletion	_	0.7
³¹ (2024)	425 nm	28.6 mW. cm ⁻²	MOF on MOF heterostructures (bidentate ligand MOF onto the UiO-66 MOF)	PEDOT: PSS	PBS +0.1 M ascorbic acid	Faradaic photoexcited electrons of bidentate ligand MO transferred from the HOMO to LUMO, migrated to the LUMO of UiO- 66 MOF	Depletion	1.56	2.50
This work	655 nm	258 mW. cm ⁻²	1) p(C ₆ NDI-T)/ gold gate 2) P-90/ gold gate 3)BBL/ gold gate 4) gAIID-2FT / gold gate	1) p(C ₆ NDI-T) 2) P-90 3) BBL 4) gAIID-2FT	PBS	Potentiometric photovoltage generation at the mixed conductor based gate	Enhancement	1) 32.7 2) 2.46 3) 6.42 4) 8.22	1) 39.0 2) 1.45 3) 14.1 4) 16.5

Abbreviation: ITO: indium tin oxide; MOF: metal-organic framework; NRs: nanorods; FTO: fluorine-doped tin oxide; QDs: quantum dots; AA: ascorbic acid; MEA: monoethanol amine; PDots: polymer dots; Pt: platinum; $ZnIn_2S_4$: zinc indium sulfide; TiO_2 : titanium oxide; OEG-SP: oligo(ethylene glycol)-spiropyran

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