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Supplementary information

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Electrolyte-gated transistors for enhanced performance bioelectronics

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Supplementary Table 1 | **Properties of EGT channel ion permeable and ion impermeable semiconductors.**

Conjugated Polyelectrolytes

OECT Material	μ (cm^2 V^{-1} s^{-1})	C^* (F cm^{-3})	[μC^*] (F cm^{-1} V^{-1} s^{-1})	Ref.
PEDOT-S^a	-	-	-	[1]
PTEBS^a	-	-	-	[1]
P3CPT	-	-	-	[2]
PTHS + EG	0.0013	124	0.16	[3,4]
CPE-K	-	134	-	[5]

Conjugated Polymer Composites

OECT Material	μ (cm^2 V^{-1} s^{-1})	C^* (F cm^{-3})	[μC^*] (F cm^{-1} V^{-1} s^{-1})	Ref.
PEDOT:PSS + EG	1.9	3.9	47	[4,6]
PEDOT:PSS + EG	-	31	100	[7]
PEDOT:PSS + H_2SO_4	-	113	490	[7]
PEDOT:PSS + Acetone (microfiber)	2.42-7.34	64	174-445	[8]
PEDOT:PSS + Acetone + H_2SO_4 (microfiber)	4.22-12.86	122	549-1500	[8]
PEDOT:TOS	0.93	136	72	[9]
PEDOT:PSTFSILi100	0.23	26	20	[9,10]
PEDOT:DS + EG	0.0064	65	2.2	[9,11]
PEDOT:PMATFSILi80	0.0024	27	0.15	[9,10]

Conjugated Polymers (p-type)

OECT Material	μ (cm^2 V^{-1} s^{-1})	C^* (F cm^{-3})	[μC^*] (F cm^{-1} V^{-1} s^{-1})	Ref.
p(g2T2-T)	~0.0001	8	9	[12]
p(g3T2-T)	0.16	211	135	[12,13]
p(g4T2-T)	0.06	192	54	[12]
p(g6T2-T)	-	-	-	[12]
p(g2T-TT)	0.94	241	261	[3,4]
gBDT-g2T	0.018	77	4.8	[13]
p(g3T2)	0.90	156	161	[14]
p(g2T2-g4T2)	1.72	187	522	[14]
p(g1T2-g5T2)	2.61	133	496	[14]
p(g0T2-g6T2)	2.95	74	302	[14]
p(gDPP-T2)	1.55	196	342	[15]
p(gDPP-TT)	0.57	184	125	[15]
p(gDPP-MeOT2)	0.28	169	57	[15]
p(gPyDPP-MeOT2)	0.03	60	-	[16]
P3MEEMT	0.19	175	53	[17]

Conjugated Polymers (n-type)

OECT Material	μ (cm^2 V^{-1} s^{-1})	C^* (F cm^{-3})	[μC^*] (F cm^{-1} V^{-1} s^{-1})	Ref.
BBL	0.0007	930	-	[2]
p(gNDI-gT2)	0.00031	397	0.18	[4,18]
p(C3-gNDI-gT2)	-	72	0.13	[19]
p(C6-gNDI-gT2)	-	59	0.16	[19]
PgNaN	0.0065	100	0.662	[20]
PgNgN	0.00019	239	0.037	[20]

Organic Semiconductors

EGOFET Material	μ ($cm^2 V^{-1} s^{-1}$)	C' ($F cm^{-2}$)	Ref.
P3HT	~0.1	$3 – 6\ 10^{-6}$	[21]
pBTTT-C14	~0.7	$1 – 5\ 10^{-6}$	[22,23]
DPP-DTT	~7.5	$7\ 10^{-6}$	[24,25]
Pentacene	~0.5	$7.8\ 10^{-6}$	[26]
α6T	~0.4	$2\ 10^{-6}$	[27]
DDFTTF	~0.2	-	[28]

(μ, electronic charge carrier mobility; C*, capacitance per univ volume. ^{a}A polymer-in-salt electrolyte was employed rather than the common aqueous 0.1 M sodium chloride solution.

Supplementary Table 2 | **Key figures of merit for electrolyte gated transistors with inorganic semiconductor channels.** μ is the electronic mobility, C_G is the gate capacitance per unit area, V_T is the threshold voltage, g_m the maximum transconductance and W the channel width.

Semiconductor	Electrolyte	μ ($cm^2 V^{-1} s^{-1}$)	C_G (μF cm^{-2})	I_{ON}/I_{OFF}	V_T (V)	g_m/W (S m^{-1})	Ref.
a-$InGaZn_2O_5$ (a-IGZO)	-	3.4	-	10^3	-	-	29
	Deionized water	-	-	1.4 10^3	-0.41	-	30
	Hydrated bovine serum albumin	116.9 ± 4.6	1.3	1.8 10^5	0.25	-	31
	Polymer [polyethylene oxide (PEO) + $LiClO_4$]	42	30	10^4 - 10^5	0.7-0.8	-	32
	Composite solid polymer electrolyte (CSPE)	>100	10	10^6- 10^7	0.2 – -0.2	>30	33
In_2O_3	CSPE	5	23 – 48	-	0.07 (top-gate) 0.26 (side-gate)	-	34,35
	CSPE	-	(5 – 30) 10^{-3}	~ 10^5	0.01 – -0.27	-	36
	SPE	0.26	~ 3.2	2 10^3	0.54	-	37
	CSPE	>5	7.6	>2 10^4	-0.22	3.12	34
	Ion gel	-	5.4	1.3 10^6	~ -0.2	-	38
	CSPE	16	10 – 42	~ 10^5	0.23	-	39
ZnO	0.1 M KCl	0.81	8.04	10^3	0.57	0.107	40
	Ion gel	2 – 9	24	>10^5	0.5	19	41
	Ion gel	1.61 ± 0.09	3.80	2.15 10^5	0.9 ± 0.05	-	42
	Ion gel	2.06	2-31	≥ 10^4	< 2	-	43
Graphene	Phthalate buffer, Phosphate buffer solution (PBS), and borate buffer	1000 (hole) 2000 (electron)	2	-	~ 0.04	3.6	44
	Sodium phosphate buffer	1700 (hole)	2	-	0.43	4.23 10^{-3} S/V	45
Carbon nanotubes	NaCl	1000 - 4000	7 10^{-5}	-	-	7 10^3	46
	Dulbecco's modified Eagle's medium (DMEM)	-	-	-	~ -0.4	5.5 10^{-3}	47
	Ion gel	9 ± 4 (hole) 3 ± 1 (electron)	-	3 10^5 2 10^5	-0.53 0.66	-	48
	Polyfluorinated electrolyte	11.5 (hole) 14.7 (electron)	10	>10^5	-0.32 0.68	2	49
	Ion gel	> 10	-	> 10^3	-	-	50
MoS_2	Ionic liquid ([EMIM] [TFSI])	~ 0.16	1.9 F cm^{-3}	10^2	~0.2	-	51
	PBS	13.5	-	10^7	-0.29	-	52
	PMMA/$LiClO_4$/PC/EC	-	10	10^4	< 1	-	53
	Ion gel	9	5.9	10^7	0.2	-	54
	PBS	26	5.2	10^4	0.11	1.98	55
WS_2	Ionic liquid ([EMIM] [TFSI])	0.01	30 F/cm^3	> 10^4	1.0 ± 0.1	0.27	56
ITO	Starch-based solid electrolyte	14.9	1.6	1.9 10^7	~0.2	-	57
Tellurene	$LiClO_4$/PEO	~ 500	-	10^5 – 10^6		-	58

Supplementary Table 3 | **Strategies for direct grafting of linkers to the gate electrode.**

Linker type	Interaction	Methodology
Polyhistidine Tag (synthetic hexamer of histidine).[59,60]	- Electrostatic interactions (image potential, p-stacking). - Coordination binding to divalent cations (Cu^{2+}, Ni^{2+}) chelated by surface bound molecules.	To graft an engineered protein bearing the Polyhistidine Tag to the metal surface. This approach is most effective for Ni and Au electrodes, albeit can be extended to any metal surface.
Thiolated flexible linkers.[61]	Metal-Sulphur (M-S) covalent bond between metal atoms and thiol.	To directly bind oligonucleotides and aptamers to metal electrodes.
Naturally occurring or engineered cysteine.[23,62,63]	M-S covalent bond between surface atoms and cysteine.	To graft native or mutant proteins (including antibodies and enzymes) by exploiting exposed cysteine(s).
Amines [64]**, aryldiazonium salts** [25]	Covalent bond between the surface and either amine nitrogen or the aryl group.	To electrograft typically low molecular weight probes such as oligopeptides on different substrates[65] either through oxidative (amines) or reductive (diazonium salts) electrografting.
Alkanethiol SAMs terminated with **a) carboxylic acid, or** **b) amino group.**	- M-S covalent bond; - Amide bond with proteins.	This methodology requires two steps: i) Chemical activation by EDC/NHS reaction of -COOH group either as SAM terminal group or the protein C-terminus; ii) Constructive assembly of a protein monolayer on SAM by formation of amide bond with the activated COOH.
Alkanethiol SAMs terminated with hydrophilic group.[62]	- M-S covalent bond; - Hydrogen bonds. alternatively, - Condensation reaction.	This strategy has been used in EGTs only for passivation or redox/pH switching. It could be exploited to graft a second monolayer by either i) hydrophilic (non-covalent) interactions, or ii) condensation, forming ester or siloxane bonds.
Alkanethiol SAMs terminated with hydrophobic groups.	- M-S covalent bond; - Hydrophobic interactions. - Van der Waals interactions.	This strategy has not been explored yet in EGT sensors. It could be exploited to assemble lipid membranes (mono- or bilayer) intercalated with proteins and enzymes.
Binary SAMs[66,67]	- Exchange of one of the two components with a thiolated linker. - Formation of amide bond with proteins with EDC/NHS.	Examples include mercaptoundecanoic acid (MUA) mixed with mercapto-heaxanoic (MHA) or -propionic acid (MPA). Biorecognition group is built on MUA, while the shorter MHA or MPA reorganizes at multiple length scales upon recognition. This “domino effect” amplifies the signal change due to local binding events.

SAMs that chemically bind the target molecules.[68]	Formation of covalent bonds between SAM and analyte.	This strategy is effective for homolog family, like catecholamines with phenylboronic acid-terminated SAM. However, the non-reversibility of the covalent bonds between probe and target leads to “poisoning” of the sensor, with consequent decrease of sensitivity and dynamic range of response.

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