Supplementary material Graphene field-effect transistors as bioanalytical sensors: design, operation and performance

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1 Literature survey

We surveyed the literature for studies on GFETs applied as bioanalytical sensors. Specifically, we selected experimental studies conducted with GFETs to detect biologically-relevant analytes, such as proteins, DNA, small molecules, ions, bacterias, viruses. We collected 85 papers fitting this criteria. For each paper, we extracted a wide range of parameters covering design, operation and performance specifications. We used "n/a" when the attribute was not applicable, and "not found" when we could not find the information in the paper. The complete database of these papers is provided online here: http://bit.ly/Beraud2020_bioGFETdatabase.

2 Analysis of reported LODs

The analysis of reported limits of detection (LoDs) was done on papers from the above collection that presented the following two critera:

- Limit of detection (LOD) explicitly stated in the paper;
- Analyte concentrations expressed in molar unit (M) or conversion possible with the information provided in the paper.

With these constraints, we collected 61 papers providing the following data:

- 20 data points from 17 papers on protein detection (Table S1),
- 14 data points from 12 papers on ions detection (Table S2),
- 10 data points from 10 papers on the detection of various small molecules (Table S3),
- 23 data points from 22 papers on DNA detection (Table S4).

For these four sets, we tabulated below the nature of the analyte, the type of graphene used, and the LOD as it is reported in the paper. In the case of DNA detection, we also report the length of the target DNA sequence. When necessary, we also report the molecular weight of the analyte used to convert the LOD in molar unit. All reference numbers are the same as in the main article.

We draw attention on two considerations:

- 1. The LODs were transcribed as reported in the original articles. Consequently, the tabulated data aggregates LODs calculated with various techniques, without validation or calibration of the methods used by the authors of the studies.
- 2. Graphene type is presented here as a proxy for graphene quality (as discussed in the main article). Mobility values were not directly used because they were not reported in enough papers, and because reported values often include contributions external to the quality of graphene itself, such as from the contacts.

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Graphene type	Analyte	Molecular weight	Reported LOD	LOD in M	Citation
rGO	Protective Antigen		1.2aM	1.2×10^{-18}	146
rGO	PSA-ACT		1.1 fM	$1.1 imes 10^{-15}$	41
rGO	BNP		100 fM	1×10^{-13}	174
rGO	IgG	150 KDa	1 ng/mL	$6.7 imes 10^{-12}$	147
rGO	IgE		43.2pM	4.3×10^{-11}	196
CVD	SARS-CoV-2 spike protein	90kDa	1fg/mL	1.1×10^{-17}	136
CVD	TSH		0.2×10^{-15}	2×10^{-16}	190
CVD	Ferritin		10fM	1×10^{-14}	116
CVD	HcG	36.7 kDa	1 pg/mL	2.7×10^{-14}	89
CVD	IL-6		618 fM	6.2×10^{-13}	139
CVD	Insulin		766 fM	7.6×10^{-13}	139
CVD	AFP	70 kDa	0.1 ng/mL	1.4×10^{-12}	115
CVD	Thrombin		2.6pM	2.6×10^{-12}	62
CVD	Poly-l-lysine		11 pM	1.1×10^{-11}	132
CVD	Insulin		35 pM	3.5×10^{-11}	189
CVD	IFN-γ		83 pM	8.3×10^{-11}	102
ME	cTn1	23.9 kDa	10 fg/mL	4.2×10^{-16}	117
ME	CCP	10 kDa	10 fg/mL	1×10^{-15}	117
ME	p24	24 kDa	100 fg/mL	4.2×10^{-15}	117
ME	HSP		100 pM	1×10^{-10}	88

Table S1: Reported data in GFET studies on protein detection (20 data points from 17 papers)

Graphene type	Analyte	Molecular weight	Reported LOD	LOD in M	Citation
rGO	Hg ²⁺		1 nM	1×10^{-9}	118
rGO	Hg ²⁺		1 nM	1×10^{-9}	38
rGO	Pb ²⁺		10 nM	1×10^{-8}	120
rGO	HPO_4^{2-}		26 nM	$2.6 imes 10^{-8}$	200
rGO	Ca ²⁺		1 µM	1×10^{-6}	38
CVD	K+		0.058pM	$5.8 imes 10^{-14}$	157
CVD	Hg ²⁺		10 pM	1×10^{-11}	123
CVD	Hg ²⁺		40 pM	4×10^{-11}	122
CVD	Pb ²⁺	207 g/mol	163.7 ng/L	7.9×10^{-10}	39
CVD	K+		1nM	1×10^{-9}	119
CVD	Pb ²⁺		2µM	2×10^{-6}	158
CVD	K+		27µM	2.7×10^{-5}	158
ME	Pb ²⁺	207 g/mol	37.5ng/L	1.8×10^{-10}	121
ME	Hg ²⁺	200.58 g/mol	0.1ppb	5×10^{-7}	87

Table S2: Reported data in GFET studies on ion detection (14 data points from 12 papers)

Graphene type	Analyte	Molecular weight	Reported LOD	LOD in M	Citation
rGO	Tobramycin		0.3 nM	3×10^{-10}	50
rGO	ATP		400nM	4×10^{-7}	106
CVD	Biotin		0.37pM	3.7×10^{-13}	100
CVD	Nalodextrone	341.401 g/mol	10 pg/mL	2.9×10^{-11}	49
CVD	OH radical		$10^{-9}M$	1×10^{-9}	52
CVD	Glucose		0.15 μM	1.5×10^{-7}	159
CVD	Glucose		0.46 µM	4.6×10^{-7}	40
CVD	Glucose		0.1 mM	1×10^{-4}	91
ME	Chlorpyrifos		1.8 fM	1.8×10^{-15}	56
ME	NO		0.3 nM	3×10^{-10}	51

Table S3: Reported data in GFET studies on small molecules detection (10 data points from 10 papers)

Graphene type	DNA length (mer)	LOD reported (M)	Citation
rGO	18	5×10^{-12}	60
rGO	22	1×10^{-13}	43
rGO	22	1×10^{-14}	161
rGO	22	2.4×10^{-9}	107
rGO	24	2×10^{-9}	134
CVD	11	2×10^{-12}	129
CVD	12	1×10^{-12}	104
CVD	12	1×10^{-11}	105
CVD	15	1×10^{-13}	83
CVD	15	1×10^{-17}	162
CVD	19	1×10^{-9}	108
CVD	20	1×10^{-11}	53
CVD	20	3×10^{-9}	135
CVD	20	1×10^{-13}	63
CVD	21	5×10^{-15}	57
CVD	22	6×10^{-19}	59
CVD	22	2×10^{-17}	59
CVD	22	1×10^{-14}	125
CVD	25	2.5×10^{-17}	81
CVD	30	1×10^{-9}	109
CVD	39	8×10^{-21}	124
CVD	60	1×10^{-15}	65
CVD	80	1×10^{-18}	58

Table S4: Reported data in GFET studies on DNA hybridization detection (23 data points from 22 papers)