

## SUPPORTING INFORMATION

# PEDOT:PSS Hydrogel Gate Electrodes for OTFT Sensors

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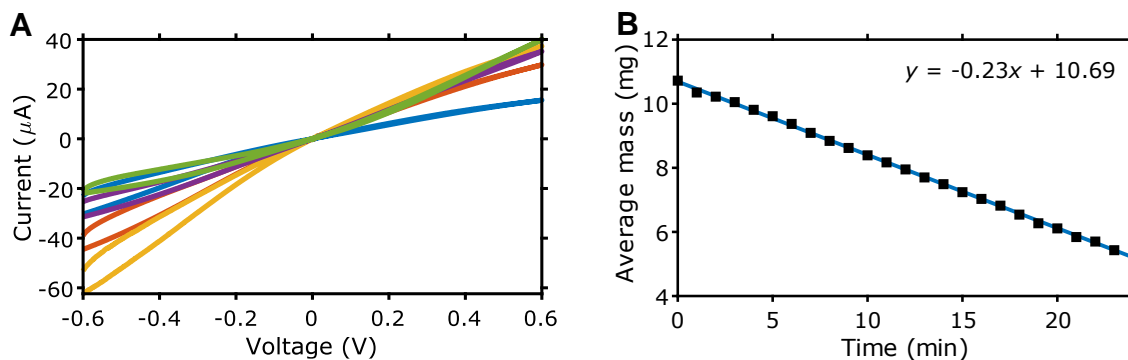
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## Figures of Merit Definitions

**Table S1:** Definitions used for extraction of HIFET figures of merit from the transfer characteristics. These definitions are adapted from the same provided in previous publications.<sup>1</sup> It is noted that non-ideal transistor characteristics can lead to systematic inaccuracies in some these estimates.

<b>ON current</b>	The maximum $I_{ds}$ in the transfer sweep (at $V_g = V_{ds} = -1$ V).
<b>OFF current</b>	The minimum $I_{ds}$ in the transfer sweep.
<b><math>V_{OFF}</math></b>	The $V_g$ at which $I_{ds}$ is at its minimum (the OFF current).
<b><math>g_m/W</math></b>	Maximum derivative of the transfer curve ( $\partial I_{ds}/\partial V_g$ ), normalised by the channel width ( $W = 3$ mm).
<b><math>\mu_{sat} \times C</math></b>	The product of saturation mobility and capacitance is calculated using the following: $\mu_{sat} \times C = \frac{2L}{W} \left( \frac{\partial \sqrt{I_{ds}}}{\partial V_g} \right)^2$ For our devices, $L = 50$ $\mu\text{m}$ and $W = 3$ mm. The value of $\partial \sqrt{I_{ds}}/\partial V_g$ is estimated by extracting the gradient of the line fitted to the most linear region of the plot of $\sqrt{I_{ds}}$ vs. $V_g$ .
<b><math>I_g</math> (ON)</b>	The $I_g$ recorded simultaneously with the ON current (when $I_{ds}$ is at its maximum).
<b><math>I_g</math> (OFF)</b>	The $I_g$ recorded simultaneously with the OFF current (when $I_{ds}$ is at its minimum).



**Figure S1:** (A) Current-Voltage characteristics of five hydrogel electrodes, showing both forward and backward sweeps. Different colours correspond to individual electrodes. (B) Average mass of 10 hydrogel gate electrodes drying in ambient conditions over 24 minutes. Blue line represents a linear fit to the data (equation displayed on plot).

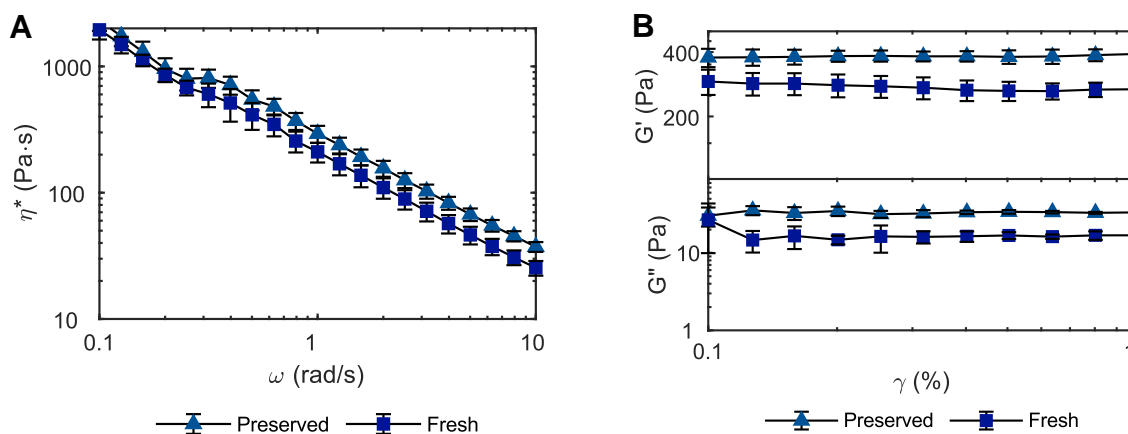
**Table S2:** Rheological figures of merit (with standard deviations) for preserved PEDOT:PSS hydrogel (24 hours) and freshly made PEDOT: PSS hydrogel. Averages were taken from multiple measurements of two samples under the same conditions.

Hydrogel	$\eta^*_{a}/\eta^*_{b}$	$G'$ (Pa) <sup>c</sup>	$G''$ (Pa) <sup>c</sup>	$\tan\delta^c$
Preserved	61.8±5.6	379.9±27.6	33.9±1.8	0.089±0.055
Fresh	76.6±10.2	263.3±27.1	16.8±1.7	0.064±0.103

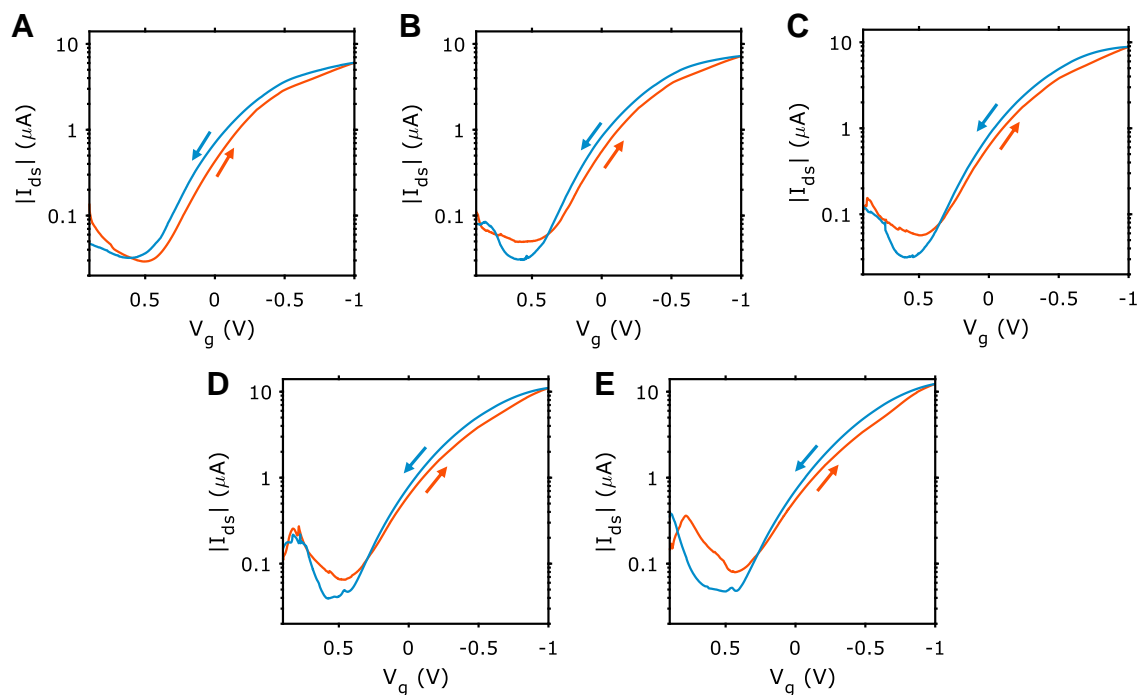
<sup>a</sup> Complex viscosity measured at 0.1 rad/s.

<sup>b</sup> Complex viscosity measured at 10 rad/s.

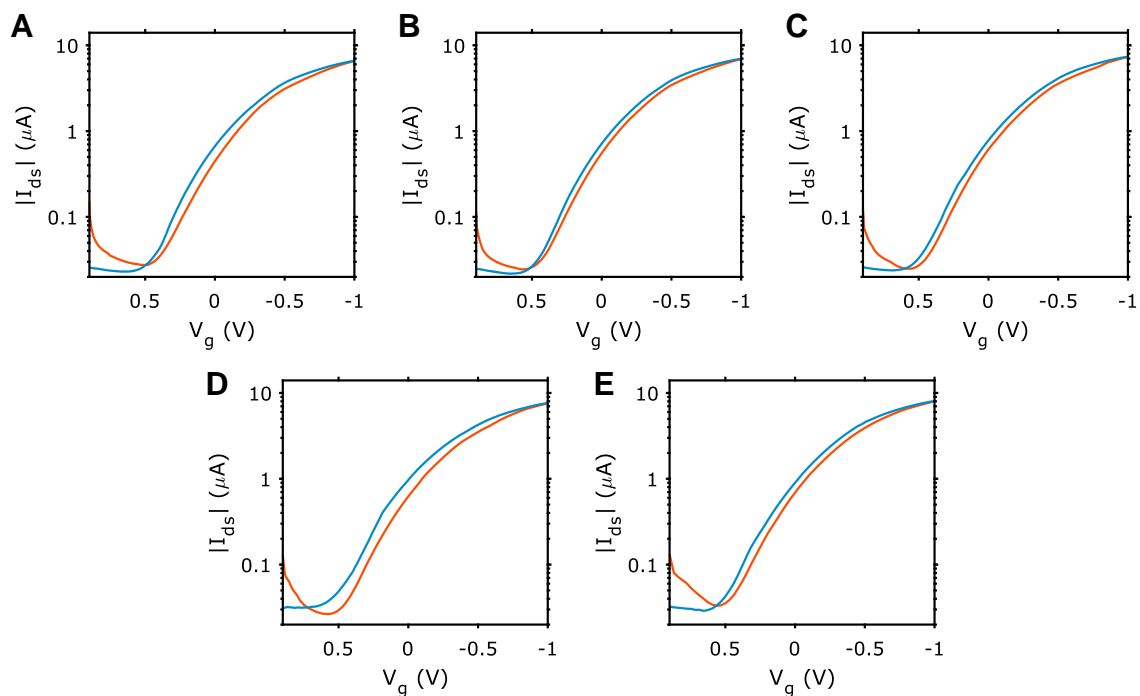
<sup>c</sup> Values recorded at 0.5% shear strain (linear region).



**Figure S2:** (A) The variation in complex viscosity ( $\eta^*$ ) with angular frequency ( $\omega$ ) for fresh and preserved (stored in water for 24 hours) hydrogel samples. (B) Plot of storage ( $G'$ ) and loss ( $G''$ ) moduli with shear strain ( $\gamma$ ), showing results for both fresh and preserved hydrogel samples. Each data point shown represents an average of 3 measurements of two hydrogel samples. Error bars represent one standard deviation.



**Figure S3:** (A-E) Chronological sequence of transfer characteristics for a single PEDOT:PSS hydrogel-gated HIFET. Each transfer sweep (forward and back) was separated by about 1 min 40 s. Over time, there is increasing non-ideal characteristics in the  $+V_g$  regime, and the ON current increases. Figures of merit are provided in Table S3.



**Figure S4:** Chronological sequence of transfer characteristics for a single PEDOT:PSS hydrogel-gated HIFET, at varied  $V_g$  sweep rates: (A)  $58 \text{ mV s}^{-1}$ , (B)  $86 \text{ mV s}^{-1}$ , (C)  $170 \text{ mV s}^{-1}$ , (D)  $407 \text{ mV s}^{-1}$ , and (E)  $528 \text{ mV s}^{-1}$ . Both ON and OFF currents increase with time and increasing sweep rate. Average figures of merit for HIFETs recorded at different sweep rates are given in Table S2.

**Table S3:** The average figures of merit for hydrogel-gated HIFETs measured repeatedly with progressively faster sweep rates. Sweep rate was controlled by varying the number of steps sampled per transfer sweep (from 1001 to 63). The actual sweep rate for each device was not exactly consistent, but averages are provided. Each device tested (5 in all) was initially sampled at 1001 steps, and then in order to 63 steps.

Steps	1001	501	251	126	63
Sw. Rt. (mV/s)	57.04 ± 3.32	89.01 ± 7.09	177.95 ± 37.75	398.07 ± 37.63	546.94 ± 51.45
ON/OFF	192.74 ± 80.55	205.35 ± 87.77	198.33 ± 70.06	192.12 ± 60.22	212.40 ± 62.65
ON (μA)	6.69 ± 0.35	7.25 ± 0.56	7.60 ± 0.88	10.03 ± 1.58	11.62 ± 2.36
OFF (μA)	0.04 ± 0.02	0.05 ± 0.02	0.05 ± 0.02	0.06 ± 0.03	0.06 ± 0.03
$g_m$ (μS/mm)	2.78 ± 0.72	2.90 ± 0.62	2.97 ± 0.55	4.07 ± 1.02	5.09 ± 1.80
$V_{OFF}$ (V)	0.60 ± 0.11	0.60 ± 0.09	0.67 ± 0.11	0.68 ± 0.13	0.61 ± 0.11
$\mu_{sat} \times C$ (μA/V <sup>2</sup> )	0.14 ± 0.03	0.16 ± 0.03	0.17 ± 0.04	0.19 ± 0.05	0.23 ± 0.03
$I_g$ at -1 V (μA)	-1.92 ± 2.03	-1.82 ± 1.21	-2.53 ± 1.89	-4.93 ± 4.17	-11.16 ± 11.87
$I_g$ at $V_{OFF}$ (μA)	0.06 ± 0.01	0.08 ± 0.03	0.18 ± 0.19	0.54 ± 0.46	0.60 ± 0.68

**Table S4:** In our examination of the effect of sweep rate, we could not control for the effect of time or repeated sampling on each device. So, for comparison, we also tested a single device 5 times at the same sweep rate (1001 steps samples per transfer sweep). We see that the same effect on the ON current and transconductance is evident here, showing that this is simply an effect of time and/or repeated measurement rather than sweep rate. Only the effect on the gate currents is not evident here.

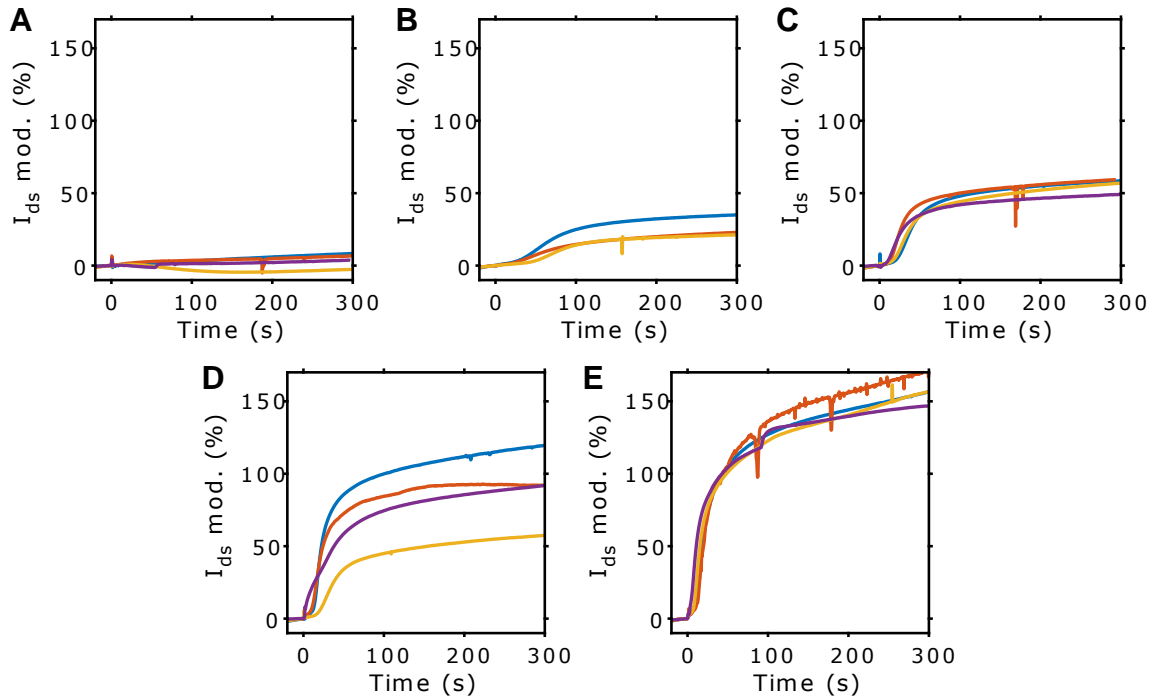
Steps	Transfer 1	Transfer 2	Transfer 3	Transfer 4	Transfer 5
Sw. Rt. (mV/s)	56.41	51.13	48.44	47.64	50.01
ON/OFF	188.09	238.16	281.14	279.55	259.07
ON (μA)	6.03	7.23	8.85	11.03	12.31
OFF (μA)	0.03	0.03	0.03	0.04	0.05
$g_m$ (μS/mm)	2.27	2.94	3.74	4.65	5.46
$V_{OFF}$ (V)	0.62	0.57	0.60	0.58	0.50
$\mu_{sat} \times C$ (μA/V <sup>2</sup> )	0.16	0.19	0.23	0.25	0.25
$I_g$ at -1 V (μA)	-1.13	-1.19	-1.24	-1.49	-1.49
$I_g$ at $V_{OFF}$ (μA)	0.08	0.07	0.07	0.08	0.09

## PEDOT:PSS hydrogel-gated HIFET characteristics

**Table S5:** Average figures of merit for a large batch (19 devices) of hydrogel-gated HIFETs.

Figure of Merit	Average $\pm$ S.D.
Sw. Rt. (mV/s)	70.07 $\pm$ 6.77
<b>ON/OFF</b>	160.48 $\pm$ 53.64
<b>ON</b> ( $\mu$ A)	6.99 $\pm$ 1.57
<b>OFF</b> ( $\mu$ A)	0.05 $\pm$ 0.02
<b><math>g_m</math></b> ( $\mu$ S/mm)	2.86 $\pm$ 0.83
<b><math>V_{OFF}</math></b> (V)	0.58 $\pm$ 0.10
<b><math>\mu_{sat} \times C</math></b> ( $\mu$ A/V <sup>2</sup> )	0.16 $\pm$ 0.04
<b><math>I_g</math> at -1 V</b> ( $\mu$ A)	-0.97 $\pm$ 1.57
<b><math>I_g</math> at <math>V_{OFF}</math></b> ( $\mu$ A)	0.09 $\pm$ 0.03

## KCl sensing results



**Figure S5:** Transient  $I_{ds}$  modulations for PEDOT:PSS hydrogel-gated HIFETs, upon deposition of 5  $\mu$ L of KCl solutions of different concentrations: **(A)** 0 M, **(B)** 0.01 M, **(C)** 0.1 M, **(D)** 1 M, and **(E)** 4 M. Sensing data for individual devices are shown as differently coloured curves. Selected representative curves are provided in Figure 4A in the main text.

**Table S6:** Summary of data from transient  $I_{ds}$  modulations for KCl sensing.

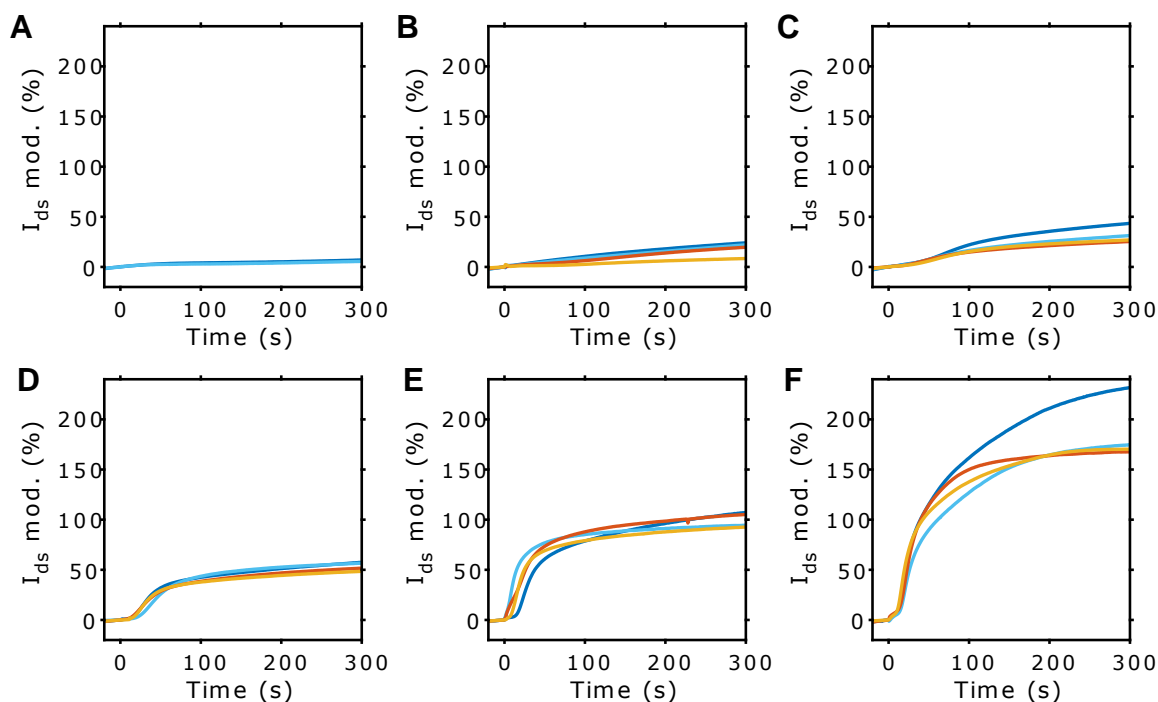
	0 M	0.01 M	0.1 M	1 M	4 M
<b>Max. <math>I_{ds}</math> mod. (%)</b>	5.08 ± 3.26	26.40 ± 7.51	55.98 ± 4.63	90.47 ± 25.46	159.05 ± 10.23
<b>Time to max. (s)</b>	154.00 ± 165.63	299.17 ± 0.29	297.00 ± 3.79	280.13 ± 38.75	288.13 ± 22.41
<b>Time to 90% max. (s)</b>	47.40 ± 80.90	63.80 ± 15.86	59.30 ± 13.93	71.30 ± 20.01	109.70 ± 43.25
<b><math>I_{ds}</math> (t=0) (<math>\mu</math>A)</b>	-2.38 ± 0.56	-2.30 ± 0.36	-2.12 ± 0.28	-2.05 ± 0.24	-2.40 ± 0.48
<b>Max. <math>I_{ds}</math> (<math>\mu</math>A)</b>	-2.49 ± 0.53	-2.89 ± 0.31	-3.30 ± 0.32	-3.89 ± 0.56	-6.20 ± 1.06

**Table S7:** Summary of figures of merit extracted from transfer sweeps recorded before and after depositing KCl analytes.

	0 M (deionised water)		0.01 M KCl		0.1 M KCl	
	Before	After	Before	After	Before	After
<b>ON/OFF</b>	164.27 ± 66.52	113.03 ± 39.24	172.79 ± 60.57	199.51 ± 70.20	145.49 ± 44.97	159.47 ± 73.19
<b>ON (<math>\mu</math>A)</b>	8.16 ± 1.86	7.63 ± 0.79	7.49 ± 0.75	10.98 ± 0.61	6.08 ± 0.86	11.08 ± 2.45
<b>OFF (<math>\mu</math>A)</b>	0.05 ± 0.01	0.07 ± 0.02	0.05 ± 0.01	0.06 ± 0.02	0.05 ± 0.03	0.09 ± 0.05
<b><math>g_m</math> (<math>\mu</math>S/mm)</b>	3.67 ± 1.13	3.30 ± 0.28	2.90 ± 0.22	4.37 ± 0.38	2.48 ± 0.38	4.73 ± 1.22
<b><math>V_{OFF}</math> (V)</b>	0.56 ± 0.09	0.53 ± 0.08	0.57 ± 0.08	0.54 ± 0.10	0.61 ± 0.17	0.55 ± 0.14
<b><math>\mu_{sat} \times C</math> (<math>\mu</math>A/V<sup>2</sup>)</b>	0.18 ± 0.04	0.17 ± 0.02	0.18 ± 0.01	0.26 ± 0.01	0.13 ± 0.04	0.25 ± 0.10
<b><math>I_g</math> at -1 V (<math>\mu</math>A)</b>	-0.51 ± 0.21	-0.75 ± 0.09	-0.55 ± 0.08	-1.02 ± 0.18	-1.04 ± 1.22	-2.07 ± 1.93
<b><math>I_g</math> at <math>V_{OFF}</math> (<math>\mu</math>A)</b>	0.07 ± 0.02	0.08 ± 0.02	0.09 ± 0.03	0.07 ± 0.01	0.10 ± 0.04	0.13 ± 0.07
<b>Sw.Rt. (mV/s)</b>	67.06 ± 5.05	59.99 ± 3.38	68.10 ± 7.64	59.43 ± 6.98	73.36 ± 7.92	58.97 ± 9.97

	1 M KCl		4 M KCl	
	Before	After	Before	After
<b>ON/OFF</b>	151.88 ± 29.30	157.86 ± 32.65	179.55 ± 55.98	227.51 ± 64.91
<b>ON (<math>\mu</math>A)</b>	6.35 ± 1.52	13.23 ± 3.61	7.29 ± 1.38	16.83 ± 3.02
<b>OFF (<math>\mu</math>A)</b>	0.04 ± 0.01	0.09 ± 0.03	0.04 ± 0.01	0.08 ± 0.01
<b><math>g_m</math> (<math>\mu</math>S/mm)</b>	2.45 ± 0.76	6.95 ± 4.52	2.94 ± 0.55	17.79 ± 8.61
<b><math>V_{OFF}</math> (V)</b>	0.58 ± 0.04	0.59 ± 0.16	0.58 ± 0.05	0.45 ± 0.09
<b><math>\mu_{sat} \times C</math> (<math>\mu</math>A/V<sup>2</sup>)</b>	0.16 ± 0.04	0.33 ± 0.13	0.17 ± 0.05	0.46 ± 0.14
<b><math>I_g</math> at -1 V (<math>\mu</math>A)</b>	-2.03 ± 2.86	-5.08 ± 2.94	-0.49 ± 0.01	-11.28 ± 1.79
<b><math>I_g</math> at <math>V_{OFF}</math> (<math>\mu</math>A)</b>	0.09 ± 0.01	0.19 ± 0.07	0.09 ± 0.02	0.22 ± 0.08
<b>Sw.Rt. (mV/s)</b>	69.34 ± 5.02	62.66 ± 5.79	71.54 ± 4.81	63.54 ± 9.19

## NaCl sensing results



**Figure S6:** Transient  $I_{ds}$  modulations for PEDOT:PSS hydrogel-gated HIFETs, upon deposition of 5  $\mu\text{L}$  of NaCl solutions of different concentrations: **(A)** 0 M, **(B)** 0.001 M, **(C)** 0.01 M, **(D)** 0.1 M, **(E)** 1 M, **(F)** 6 M. Sensing data for individual devices are shown as differently coloured curves. Selected representative curves are provided in Figure 4E in the main text.

**Table S8:** Summary of data from transient  $I_{ds}$  modulations for KCl sensing.

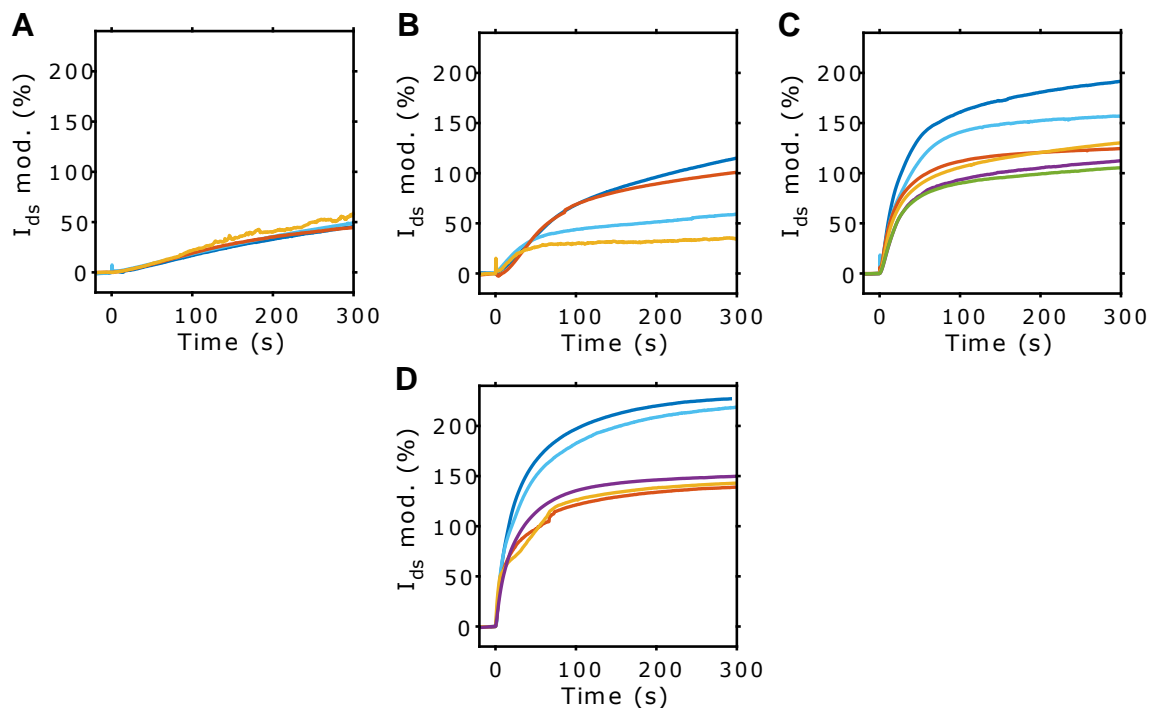
	0 M	0.001 M	0.01 M	0.1 M	1 M	6 M
<b>Max. <math>I_{ds}</math> mod. (%)</b>	$6.25 \pm 0.98$	$18.50 \pm 6.98$	$31.74 \pm 8.19$	$53.50 \pm 4.25$	$99.84 \pm 7.36$	$186.05 \pm 30.48$
<b>Time to max. (s)</b>	$299.50 \pm 0.00$	$299.13 \pm 0.75$	$299.25 \pm 0.29$	$298.75 \pm 0.87$	$298.75 \pm 1.19$	$298.13 \pm 1.18$
<b>Time to 90% max. (s)</b>	$5.75 \pm 4.40$	$85.88 \pm 57.31$	$107.63 \pm 25.82$	$82.88 \pm 11.40$	$83.00 \pm 40.06$	$123.20 \pm 32.46$
<b><math>I_{ds}(t=0)</math> (<math>\mu\text{A}</math>)</b>	$-2.66 \pm 0.05$	$-2.59 \pm 0.38$	$-2.44 \pm 0.45$	$-2.69 \pm 0.41$	$-2.91 \pm 0.28$	$-2.34 \pm 0.25$
<b>Max. <math>I_{ds}</math> (<math>\mu\text{A}</math>)</b>	$-2.83 \pm 0.03$	$-3.05 \pm 0.25$	$-3.19 \pm 0.46$	$-4.11 \pm 0.53$	$-5.80 \pm 0.42$	$-6.64 \pm 0.57$

**Table S9:** Summary of figures of merit extracted from transfer sweeps recorded before and after depositing NaCl analytes.

	0 M (deionised water)		0.001 M NaCl		0.01 M NaCl	
	Before	After	Before	After	Before	After
<b>ON/OFF</b>	231.45 ± 68.42	172.75 ± 47.13	138.69 ± 69.36	98.25 ± 33.65	89.98 ± 69.61	78.80 ± 40.07
<b>ON</b> (µA)	6.77 ± 1.45	6.95 ± 0.54	6.42 ± 0.56	6.94 ± 0.48	6.50 ± 1.75	9.01 ± 1.69
<b>OFF</b> (µA)	0.03 ± 0.00	0.04 ± 0.01	0.06 ± 0.03	0.08 ± 0.03	0.13 ± 0.09	0.15 ± 0.08
<b>g<sub>m</sub></b> (µS/mm)	2.65 ± 0.59	2.54 ± 0.34	2.50 ± 0.19	2.40 ± 0.30	3.43 ± 1.33	3.68 ± 1.09
<b>V<sub>OFF</sub></b> (V)	0.70 ± 0.01	0.63 ± 0.00	0.62 ± 0.12	0.54 ± 0.07	0.50 ± 0.16	0.44 ± 0.09
<b>µ<sub>sat</sub>×C</b> (µA/V <sup>2</sup> )	0.17 ± 0.03	0.14 ± 0.01	0.15 ± 0.02	0.15 ± 0.02	0.15 ± 0.04	0.21 ± 0.04
<b>I<sub>g</sub> at -1 V</b> (µA)	-1.02 ± 0.52	-1.50 ± 0.62	-0.38 ± 0.30	-0.67 ± 0.20	-0.39 ± 0.30	-1.04 ± 0.23
<b>I<sub>g</sub> at V<sub>OFF</sub></b> (µA)	0.05 ± 0.00	0.07 ± 0.01	0.11 ± 0.10	0.13 ± 0.10	0.14 ± 0.06	0.15 ± 0.06
<b>Sw.Rt.</b> (mV/s)	70.05 ± 1.19	67.55 ± 2.48	65.38 ± 15.73	66.63 ± 14.85	74.89 ± 11.06	72.36 ± 8.12

	0.1 M NaCl		1 M NaCl		6 M NaCl	
	Before	After	Before	After	Before	After
<b>ON/OFF</b>	102.59 ± 108.58	112.55 ± 90.27	104.25 ± 72.14	105.87 ± 70.42	123.81 ± 93.69	145.75 ± 48.52
<b>ON</b> (µA)	6.70 ± 1.00	13.27 ± 0.65	7.70 ± 1.73	14.86 ± 1.48	6.32 ± 1.10	16.95 ± 1.26
<b>OFF</b> (µA)	0.17 ± 0.12	0.18 ± 0.08	0.13 ± 0.11	0.25 ± 0.18	0.08 ± 0.04	0.13 ± 0.05
<b>g<sub>m</sub></b> (µS/mm)	2.74 ± 0.52	5.50 ± 0.68	3.05 ± 0.79	9.17 ± 2.18	2.52 ± 0.44	16.09 ± 2.06
<b>V<sub>OFF</sub></b> (V)	0.49 ± 0.18	0.49 ± 0.11	0.58 ± 0.15	0.58 ± 0.10	0.56 ± 0.12	0.37 ± 0.18
<b>µ<sub>sat</sub>×C</b> (µA/V <sup>2</sup> )	0.18 ± 0.04	0.29 ± 0.02	0.19 ± 0.04	0.30 ± 0.03	0.15 ± 0.03	0.57 ± 0.15
<b>I<sub>g</sub> at -1 V</b> (µA)	-0.54 ± 0.61	-1.65 ± 0.74	-0.28 ± 0.23	-3.87 ± 0.95	-0.19 ± 0.09	-7.34 ± 1.74
<b>I<sub>g</sub> at V<sub>OFF</sub></b> (µA)	0.13 ± 0.06	0.19 ± 0.05	0.13 ± 0.08	0.29 ± 0.09	0.09 ± 0.04	0.25 ± 0.06
<b>Sw.Rt.</b> (mV/s)	76.03 ± 13.04	69.14 ± 5.60	76.11 ± 11.54	74.93 ± 7.35	67.72 ± 7.46	73.70 ± 6.63



**H<sub>2</sub>O<sub>2</sub> sensing results**

**Figure S7:** Transient  $I_{ds}$  modulations for PEDOT:PSS hydrogel-gated HIFETs, upon deposition of 5  $\mu\text{L}$  of  $\text{H}_2\text{O}_2$  solutions of different concentrations: **(A)** 0.03%, **(B)** 0.3%, **(C)** 3%, **(D)** 30%. Sensing data for individual devices are shown as differently coloured curves.

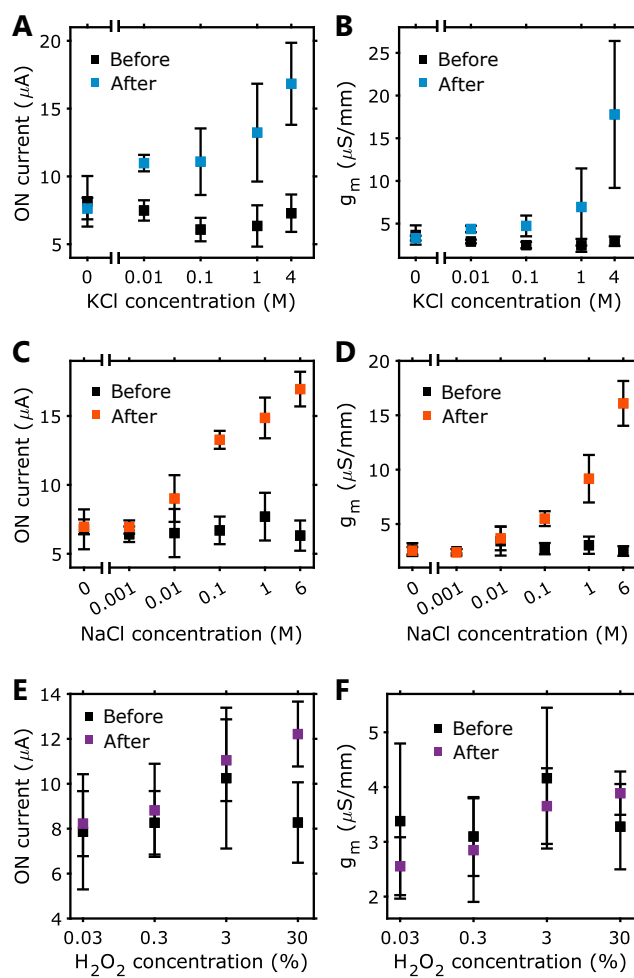
**Table S10:** Summary of data from transient  $I_{ds}$  modulations for  $\text{H}_2\text{O}_2$  sensing.

	0.03%	0.3%	3%	30%
<b>Max. <math>I_{ds}</math> mod. (%)</b>	49.59 $\pm$ 6.53	77.65 $\pm$ 36.75	136.87 $\pm$ 32.17	175.53 $\pm$ 43.55
<b>Time to 90% max. (s)</b>	147.42 $\pm$ 69.09	106.33 $\pm$ 55.12	95.93 $\pm$ 31.52	87.92 $\pm$ 18.63
<b><math>I_{ds}</math> (t=0) (<math>\mu\text{A}</math>)</b>	-2.58 $\pm$ 0.42	-2.45 $\pm$ 0.39	-2.55 $\pm$ 0.79	-2.44 $\pm$ 0.68
<b>Max. <math>I_{ds}</math> (<math>\mu\text{A}</math>)</b>	-3.86 $\pm$ 0.68	-4.28 $\pm$ 0.75	-5.83 $\pm$ 1.21	-6.49 $\pm$ 0.97

**Table S11:** Summary of figures of merit extracted from transfer sweeps recorded before and after depositing H<sub>2</sub>O<sub>2</sub> analytes.

	0.03%		0.3%		3%	
	Before	After	Before	After	Before	After
<b>ON/OFF</b>	69.24 ± 47.39	51.07 ± 32.65	89.42 ± 65.23	67.16 ± 66.15	103.72 ± 59.35	47.90 ± 24.38
<b>ON</b> (μA)	7.86 ± 2.57	8.22 ± 1.45	8.26 ± 1.41	8.82 ± 2.07	10.25 ± 3.14	11.05 ± 1.82
<b>OFF</b> (μA)	0.21 ± 0.19	0.32 ± 0.31	0.14 ± 0.07	0.49 ± 0.55	0.11 ± 0.04	0.28 ± 0.11
<b>g<sub>m</sub></b> (μS/mm)	3.38 ± 1.42	2.56 ± 0.53	3.10 ± 0.72	2.85 ± 0.95	4.16 ± 1.29	3.65 ± 0.69
<b>V<sub>OFF</sub></b> (V)	0.51 ± 0.15	0.59 ± 0.15	0.57 ± 0.17	<b>0.67</b> ± 0.26	0.47 ± 0.12	0.80 ± 0.08
<b>μ<sub>sat</sub>×C</b> (μA/V <sup>2</sup> )	0.18 ± 0.06	0.14 ± 0.04	0.18 ± 0.04	0.13 ± <b>0.08</b>	0.24 ± 0.08	0.21 ± 0.04
<b>I<sub>g</sub> at -1 V</b> (μA)	-7.62 ± 7.62	-12.29 ± 13.24	-7.79 ± 10.26	-12.58 ± 16.64	-0.36 ± 0.23	-1.05 ± 0.20
<b>I<sub>g</sub> at V<sub>OFF</sub></b> (μA)	0.35 ± 0.26	0.62 ± 0.59	0.19 ± 0.08	0.53 ± 0.41	0.14 ± 0.06	0.37 ± 0.10
<b>Sw.Rt.</b> (mV/s)	90.12 ± 4.41	93.61 ± 11.31	85.27 ± 11.38	91.60 ± 17.16	75.47 ± 7.43	90.25 ± 4.97

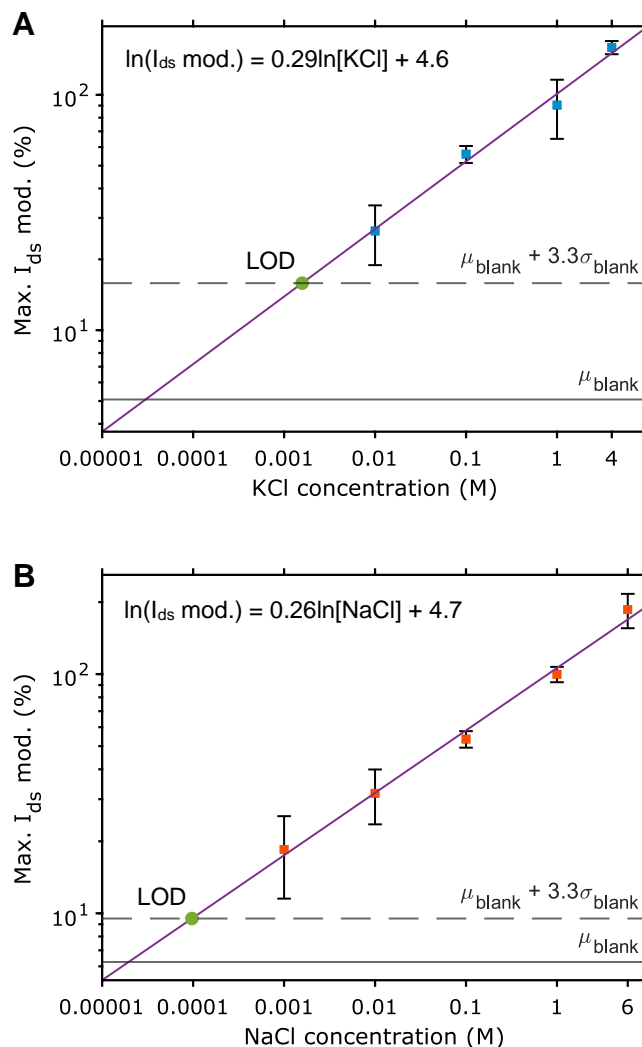
	30%	
	Before	After
<b>ON/OFF</b>	151.32 ± 82.32	83.84 ± 58.87
<b>ON</b> (μA)	8.27 ± 1.79	12.21 ± 1.44
<b>OFF</b> (μA)	0.09 ± 0.07	0.63 ± 0.93
<b>g<sub>m</sub></b> (μS/mm)	3.28 ± 0.78	3.89 ± 0.40
<b>V<sub>OFF</sub></b> (V)	0.57 ± 0.20	0.72 ± 0.24
<b>μ<sub>sat</sub>×C</b> (μA/V <sup>2</sup> )	0.19 ± 0.04	0.20 ± 0.05
<b>I<sub>g</sub> at -1 V</b> (μA)	-5.24 ± 6.40	-4.56 ± 5.21
<b>I<sub>g</sub> at V<sub>OFF</sub></b> (μA)	0.13 ± 0.07	0.82 ± 0.86
<b>Sw.Rt.</b> (mV/s)	79.47 ± 12.91	97.86 ± 12.95



**Figure S8:** Average ON currents and transconductances ( $g_m$ ) of HIFETs before and after depositing analyte solutions of various concentrations: **(A,B)** KCl, **(C,D)** NaCl, and **(E,F)**  $\text{H}_2\text{O}_2$ . Figure 4 in the main paper plots the same data, but as relative changes (modulations).

## Ion sensitivity of HIFETs and limit of detection (LOD)

If plotted on a log-log scale, the  $I_{ds}$  modulation calibration plots are approximately linear, as shown below. By fitting a linear model, we can construct calibration equations for the KCl and NaCl sensitivity of hydrogel-gated HIFETs.



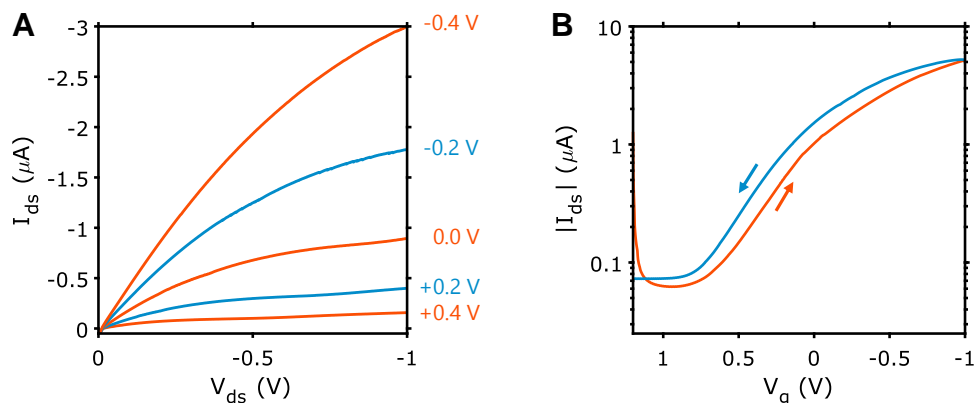
**Figure S9:** Sensing calibration curves for **(A)** KCl and **(B)** NaCl. Purple lines show fitted linear models (equations displayed). Limits of detection (LOD) are indicated by a green point. Horizontal lines show the average  $I_{ds}$  modulations for deionised water ( $\mu_{\text{blank}}$ ) and dashed lines show the upper confidence limit ( $\mu_{\text{blank}} + 3.3\sigma_{\text{blank}}$ ).

The “limit of detection” (LOD) can be defined roughly as the ion concentration predicted by the calibration equation for an  $I_{ds}$  modulation corresponding to that of the average blank sample (i.e. deionised water) plus 3.3 times the standard deviation (95% confidence interval).<sup>2</sup>

According to this definition. The limits of detection for KCl and NaCl sensitivity are as follows:

$$\text{LOD}_{\text{KCl}} = 0.0016 \text{ M} = 1.6 \text{ mM}$$

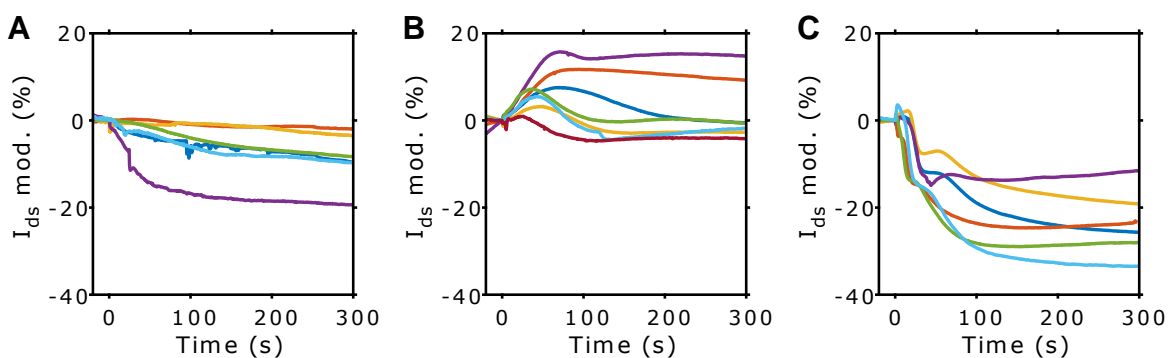
$$\text{LOD}_{\text{NaCl}} = 0.000097 \text{ M} = 97 \text{ }\mu\text{M}$$

Hydrogel-gated HIFETs with K<sup>+</sup> selective membranes

**Figure S10:** Representative (A) output and (B) transfer characteristics for a hydrogel-gated HIFET with a K<sup>+</sup> selective membrane, inserted between the PVP layer and the gate electrode.

**Table S12:** Average figures of merit for a batch (16 devices) of hydrogel-gated HIFETs with K<sup>+</sup> selective membranes.

Figure of Merit	Average $\pm$ S.D.
<b>ON/OFF</b>	72.19 $\pm$ 13.70
<b>ON (<math>\mu\text{A}</math>)</b>	5.67 $\pm$ 1.11
<b>OFF (<math>\mu\text{A}</math>)</b>	0.08 $\pm$ 0.01
<b><math>g_m</math> (<math>\mu\text{S}/\text{mm}</math>)</b>	1.87 $\pm$ 0.37
<b><math>V_{\text{OFF}}^*</math> (V)</b>	1.18 $\pm$ 0.03
<b><math>\mu_{\text{sat}} \times C</math> (<math>\mu\text{A}/\text{V}^2</math>)</b>	0.10 $\pm$ 0.02
<b><math>I_g</math> at -1 V (<math>\mu\text{A}</math>)</b>	-0.19 $\pm$ 0.04
<b><math>I_g</math> at <math>V_{\text{OFF}}</math> (<math>\mu\text{A}</math>)</b>	0.06 $\pm$ 0.03
<b>Sw. Rt. (mV/s)</b>	81.02 $\pm$ 3.26



**Figure S11:** Transient  $I_{ds}$  modulations for PEDOT:PSS hydrogel-gated HIFETs with K<sup>+</sup> selective membranes, upon deposition of 5  $\mu\text{L}$  of different analytes: (A) deionised water, (B) 1 M NaCl, (C) 1 M KCl. Sensing data for individual devices are shown as differently coloured curves.

## References

1. J. N. Arthur and S. D. Yambem, *Adv. Mater. Technol.*, 2021, 2101149.
2. D. A. Armbruster and T. Pry, *Clin. Biochem. Rev.*, 2008, 29(Suppl 1), S49-S52.