Electronic Supplementary Material (ESI) for Analytical Methods

Electrochemical DNA-Based Sensor for Simultaneous Detection of Hg^{2+} and Pb^{2+}

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Supplementary results and discussion

Table S1. Equivalent circuit element values for DNA films in absence and presence of Hg^{2+} [a]

	equivalent circuit elements						
	R_s ($\Omega\cdot cm^2$)	$C_{monolayer}$ ($\mu F\cdot cm^{-2}$)	R_{CT} ($\Omega\cdot cm^2$)	R_x ($\Omega\cdot cm^2$)	CPE ($\mu F\cdot cm^{-2}$)	n	ΔR_{CT} ($\Omega\cdot cm^2$)
DNA	5.3(0.1)	9.9(0.3)	2594(26)	2.8(0.2)	37.3(5.3)	0.9(0.01)	–
buffer	5.5(0.1)	10.2(0.4)	2560(23)	2.9(0.2)	28.5(1.8)	0.9(0.02)	36(2)
1×10^{-5} M	5.9(0.1)	7.9(0.1)	1186(35)	3.2(0.8)	42.4(2.1)	0.9(0.01)	1408(60)
1×10^{-6} M	6.0(0.1)	7.7(0.1)	1385(64)	3.3(0.3)	25.9(3.1)	0.9(0.01)	1209(38)
1×10^{-7} M	5.8(0.1)	8.2(0.2)	1561(25)	2.2(0.3)	16.4(2.5)	0.9(0.03)	1033(51)
1×10^{-8} M	5.6(0.1)	8.4(0.1)	1776(80)	3.1(0.4)	27.7(2.6)	0.9(0.01)	818(54)
1×10^{-9} M	5.6(0.1)	8.6(0.3)	1930(74)	3.2(0.4)	26.7(5.3)	0.9(0.03)	664(46)
1×10^{-10} M	5.6(0.1)	9.6(0.1)	2135(18)	3.9(0.3)	39.1(5.0)	0.9(0.02)	459(39)
1×10^{-11} M	5.7(0.1)	9.5(0.1)	2248(49)	2.3(0.1)	20.7(4.0)	0.9(0.03)	277(44)
1×10^{-12} M	5.6(0.1)	10.1(0.3)	2464(44)	3.5(0.2)	47.7(6.7)	0.9(0.03)	162(4)
1×10^{-13} M	5.7(0.1)	10.0(0.3)	2527(24)	3.0(0.4)	33.2(2.7)	0.9(0.02)	47(2)

[a] The values in parentheses represent the standard deviations from at least 5 electrode measurements.

Table S2. Equivalent circuit element values for DNA films in absence and presence of Pb^{2+} [b]

	equivalent circuit elements						
	R_s ($\Omega\cdot cm^2$)	$C_{monolayer}$ ($\mu F\cdot cm^{-2}$)	R_{CT} ($\Omega\cdot cm^2$)	R_x ($\Omega\cdot cm^2$)	CPE ($\mu F\cdot cm^{-2}$)	n	ΔR_{CT} ($\Omega\cdot cm^2$)
DNA	5.3(0.1)	9.9(0.3)	2594(26)	2.8(0.2)	37.3(5.3)	0.9(0.01)	–
buffer	5.5(0.1)	10.2(0.4)	2560(23)	2.9(0.2)	28.5(1.8)	0.9(0.02)	36(2)
1×10^{-5} M	5.8(0.1)	10.6(1.2)	988(30)	5.5(0.5)	58.4(2.4)	0.9(0.06)	1606(4)
1×10^{-6} M	5.7(0.2)	9.3(0.4)	1142(66)	2.4(0.1)	27.2(4.0)	0.9(0.06)	1452(40)
1×10^{-7} M	5.7(0.2)	9.2(0.4)	1338(30)	2.4(0.1)	25.8(4.8)	0.9(0.01)	1256(55)
1×10^{-8} M	5.7(0.2)	9.3(0.2)	1568(22)	4.2(0.5)	43.2(3.1)	0.9(0.01)	1026(49)
1×10^{-9} M	6.0(0.2)	9.8(0.7)	1760(45)	4.9(0.7)	36.3(0.2)	0.9(0.01)	834(28)
1×10^{-10} M	5.7(0.1)	9.8(0.4)	1907(90)	5.9(1.4)	33.3(0.2)	0.9(0.01)	687(62)
1×10^{-11} M	5.5(0.1)	10.2(0.3)	2098(72)	3.4(0.3)	49.8(3.6)	0.9(0.01)	496(46)
1×10^{-12} M	5.7(0.2)	9.1(1.1)	2254(56)	3.3(0.5)	27.9(1.7)	0.9(0.01)	340(30)
1×10^{-13} M	6.0(0.1)	9.5(0.7)	2404(50)	5.2(0.5)	37.2(2.0)	0.9(0.01)	190(24)
1×10^{-14} M	5.8(0.1)	10.1(0.6)	2548(14)	3.6(0.4)	45.5(1.6)	0.9(0.03)	44(5)

[b] The values in parentheses represent the standard deviations from at least 5 electrode measurements.

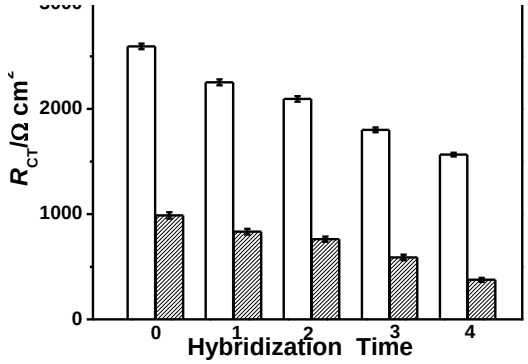

Fig.S1. Reproducibility of the assay: R_{CT} for Films of DNA (1+2+3) before (▭) and after (▨) interacting with 10^{-5} M Pb^{2+}, and further hybridization with DNA (3) (10^{-5} M DNA(3), 40 mM Tris-ClO_4, 300 mM $NaClO_4$) to form DNA (1+2+3).

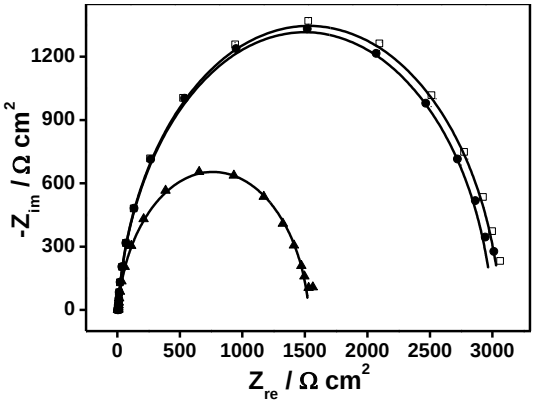

Fig. S2 Representative Nyquist plots (-Z_{im} vs Z_{re}) for films of DNA: films of DNA before (□) after incubated in cysteine + Hg^{2+} (●) and cysteine + Hg^{2+} + Pb^{2+} (▲). Concentrations: 10^{-5} M cysteine, 10^{-6} M Hg^{2+}, 10^{-6} M Pb^{2+} and 2 µM G-DNA. Measured data are shown as symbols with calculated fit to the equivalent circuit as solid lines.

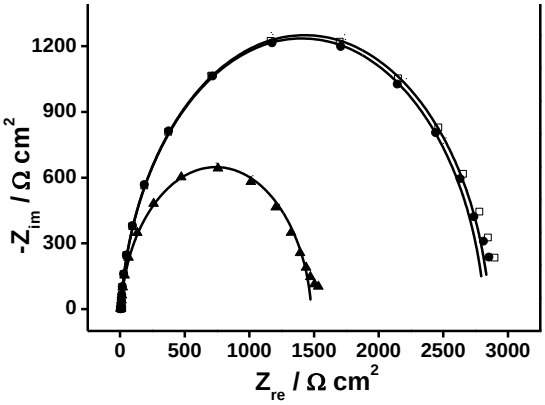

Fig. S3 Representative Nyquist plots (-Z_{im} vs Z_{re}) for films of DNA: films of DNA before (□) after incubated in G-DNA + Pb^{2+} (●) and G-DNA + Pb^{2+} + Hg^{2+} (▲). Concentrations: 10^{-5} M cysteine, 10^{-6} M Hg^{2+}, 10^{-6} M Pb^{2+} and 2 µM G-DNA. Measured data are shown as symbols with calculated fit to the equivalent circuit as solid lines.

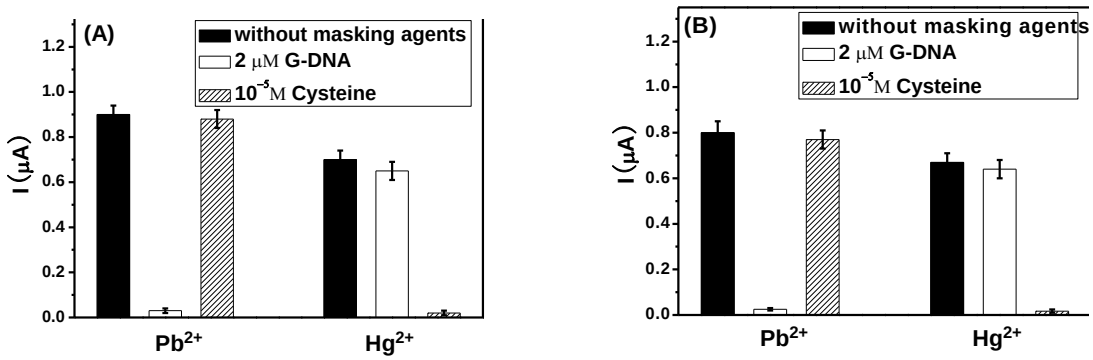

Fig. S4 Peak current (differential pulse voltammetry-DPV) of 1 mM $[Ru(NH_3)_6]^{3+}$ at DNA-modified electrodes before and after incubating with metal ions in the absence and presence of masking agents (10^{-5} M cysteine and 2 μM G-DNA): (A) 10^{-8} M Hg^{2+} and 10^{-6} M Pb^{2+} in human serum, (B) 10^{-8} M Hg^{2+} and 10^{-8} M Pb^{2+} in river water. Error bars are derived from a minimum of five electrodes. DPV scans of 1 mM $K_3[Fe(CN)_6]$ in 50 mM Tris-ClO_4 pH 7.4; pulse amplitude 100 mV, scan rate 10 mv/s, scan step 5 mV.