

Supporting Information

pH-induced on/off-switchable graphene bioelectronics

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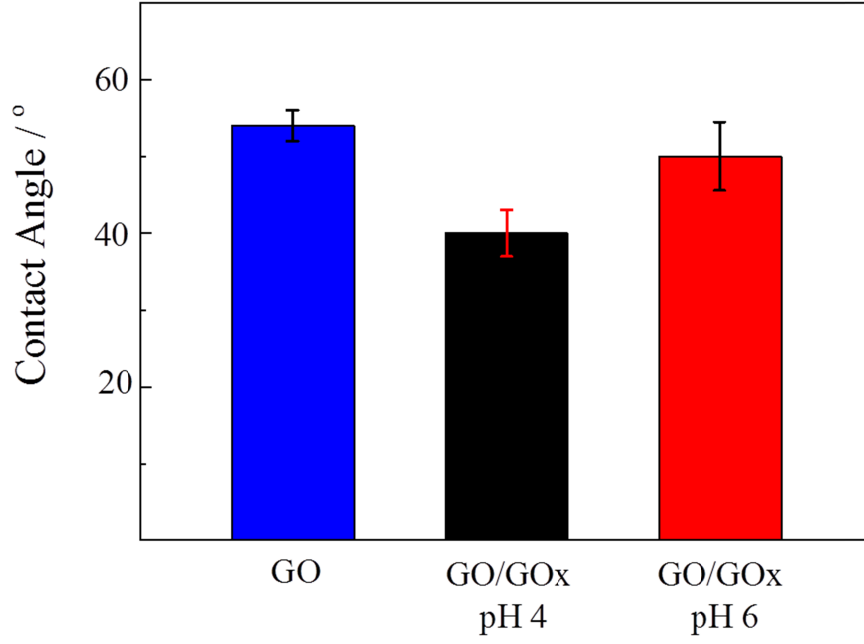
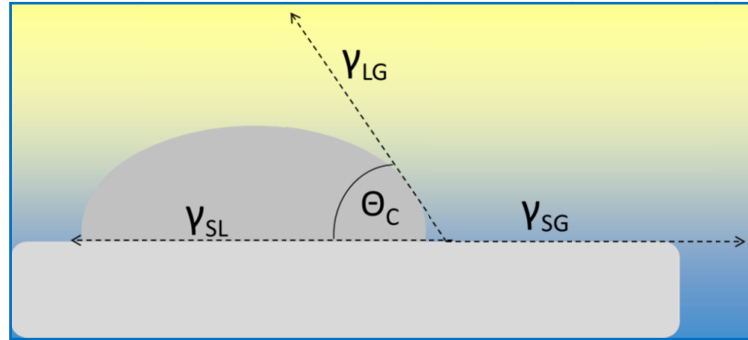


Figure S1. Water contact angles of pHGr , and GOx immobilised pHGr at pH 4 and 6.



We performed water contact angle measurement to identify the properties of surface. The contact angle is used to quantify wettability parameter of surface, which provides a thermodynamic parameter to calculate the solid interfacial tension.

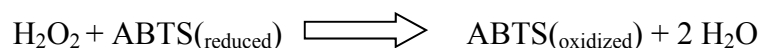
$$\cos \Theta_c = \frac{\gamma_{sg} - \gamma_{sl}}{\gamma_{lg}}$$

Young's equation which gives the measurable quantities, the contact angle Θ_c , the liquid-vapour/gas interfacial tension γ_{lg} , to the non-measurable interfacial tension γ_{sg} and γ_{sl} of the solid-vapour and solid-liquid interfaces, respectively.

The enzymatic assay was performed in order to calculate enzyme loading capacity and rate constant for enzymatic reaction. Here we describe the use of GOx coupled to horseradish peroxidase as a stable and easily measured enzyme system. The GOx is an FAD-dependent oxidoreductase which catalyses the following reaction:



In order to measure the reaction, the hydrogen peroxide produced is scavenged by HRP, which uses an electron donor to reduce further the hydrogen peroxide to water.



Here, stable, non-toxic and water soluble dye forms blue-green color when oxidized. The absorbance change was monitored at 500 nm, where the extinction coefficient is $3.6 \times 10^4 \text{ M}^{-1} \text{ cm}^{-1}$.

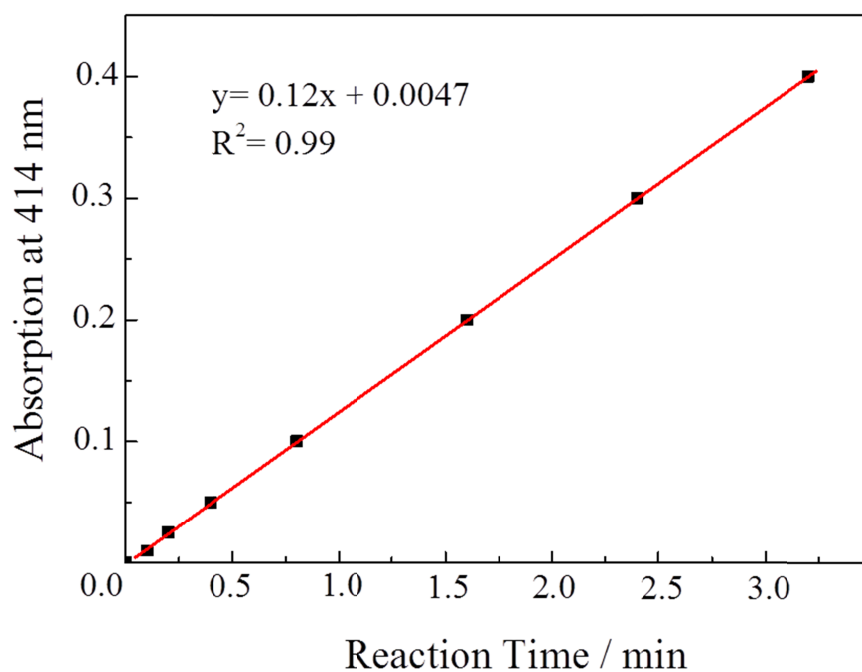


Figure S2. Time dependent absorption measurement for GOx/pHGr at pH4. (The initial rates for all samples were obtained by linear fitting of absorption vs reaction time).

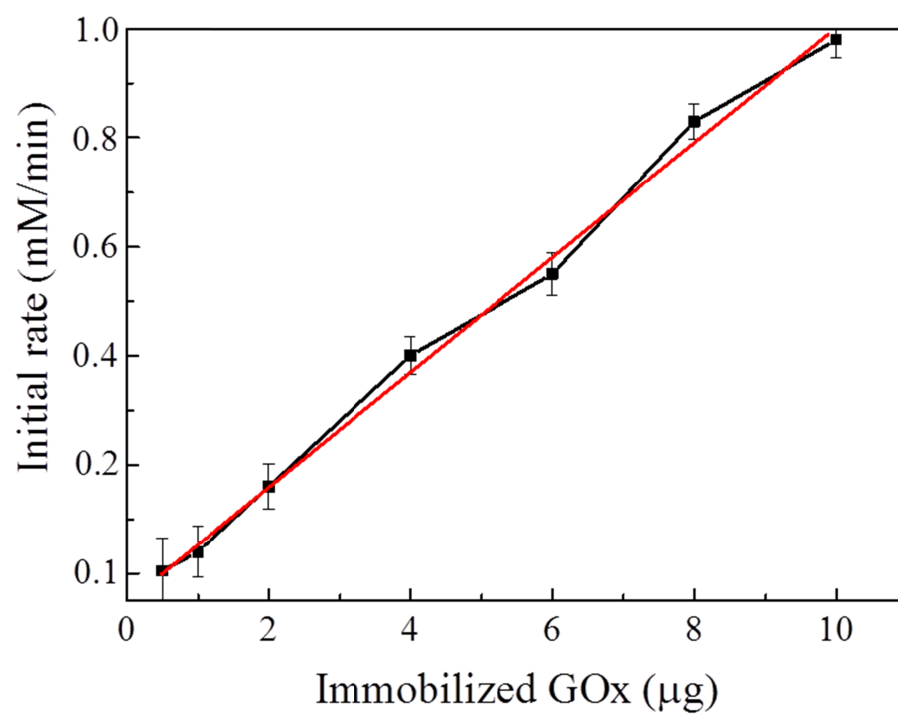


Figure S3. Initial reaction rates of GOx immobilised pHGr vs increasing amount of GOx.

Table S1. Detection limit, sensing range, sensitivity and response time of ^{pH}Gr-GOx modified GCE at pH 4 and pH 6.

Electrode	Sensitivity ^[a] [$\mu\text{A}/\mu\text{m}/\text{cm}^2$]	Detection Limit ^[b] [μM]	Dynamic Range ^[c] [mM]	Response Time ^[d] [s]
GOx/ ^{pH} Gr (pH 4)	127.57	0.012	0.01-6.0	3
GOx/ ^{pH} Gr (pH 6)	65.43	0.043	0.025-3.0	8

^[a]Sensitivities were obtained from the slopes of each calibration plots (concentration of glucose vs. current density). ^[b]Detection limits were calculated from the characteristic signal-to-noise ratio ($S/N = 3$). ^[c]Dynamic ranges were determined the linear portion of calibration curves. ^[d]Response times were determined when the currents reach to steady-state state at each addition of glucose.