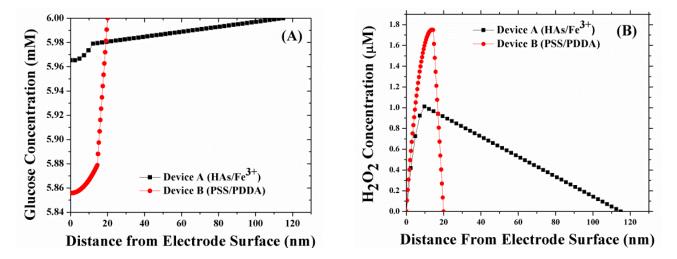
## Theoretical Analysis of the Performance of Glucose Sensors with Layer-by-Layer Assembled Outer Membranes

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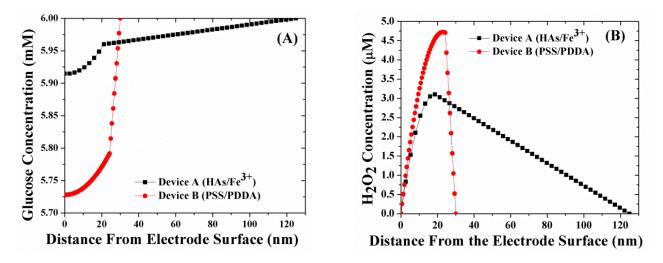
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Figures S1 and S2 show the concentration profiles of glucose and  $H_2O_2$  for Devices A and B at a  $GO_x/PPD$  thickness of 15 and 25 nm, respectively. For both these  $GO_x/PPD$  thicknesses the concentration of  $H_2O_2$  is higher in Device B (PSS/PDDA) compared to Device A (HAs/Fe<sup>3+</sup>), similar to that shown in Figure 4. This confirms that tighter (less glucose permeable) outer membranes (Device B) leads to higher build-up of  $H_2O_2$  within the sensor geometry and the phenomena is independent of the enzyme thickness. Figures 5 and 7 in the manuscript represent the time taken for the sensor to reach 90% of its saturation value. In order to obtain these values, the transient response of the sensor shown in Figures S3 was utilized. These transient responses were obtained utilizing Equation (16).

**Figure S1.** (A) Simulated glucose concentration profile in the multi-layer sensor system consisting of 15 nm  $GO_x/PPD$  as the first layer, and different LBL membranes as the second layer (HAs/Fe<sup>3+</sup> and PSS/PDDA); (B) simulated H<sub>2</sub>O<sub>2</sub> concentration profile in the multi-layer sensor system consisting of 15 nm  $GO_x/PPD$  as the first layer, and different LBL membranes as the second layer (HAs/Fe<sup>3+</sup> and PSS/PDDA).



**Figure S2.** (A) Simulated glucose concentration profile in the multi-layer sensor system consisting of 25 nm  $GO_x/PPD$  as the first layer, and different LBL membranes as the second layer (HAs/Fe<sup>3+</sup> and PSS/PDDA); (B) simulated H<sub>2</sub>O<sub>2</sub> concentration profile in the multi-layer sensor system consisting of 25 nm  $GO_x/PPD$  as the first layer, and different LBL membranes as the second layer (HAs/Fe<sup>3+</sup> and PSS/PDDA).



**Figure S3.** Simulated transient response of Devices A and B at (**A**) 2 mM, (**B**) 4 mM and (**C**) 20 mM glucose.

