pH and ion sensitivity of flexible graphene transistors for physiological applications

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Abstract
Graphene is a perfectly suited material for sensing applications in physiological media. It is biocompatible, flexible and has a wide electrochemical window. Due to its 2D-nature, graphene has the highest surface to volume ratio possible, which makes it very sensitive to the presence of charge at its surface. It also has an interfacial capacitance weakly dependent on the ionic composition since it is dominated by the quantum capacitance near the Dirac point. Moreover, graphene has field-effect behaviour which allows it to be used as an active material in transistors\textsuperscript{[1]}. Although the pH and ion sensitivities of graphene transistors have been already reported \textsuperscript{[2]}, their interaction has not been exhaustively described. Here, we use a Freundlich adsorption isotherm to model the pH sensitivity originating from acidic groups at graphene surface, and the Grahame equation to model the ionic charge in the diffusive layer. Combining both equations, the variation of the charge neutrality point (CNP) is obtained. Simulations are performed for different pH and concentration of monovalent and divalent ions (Na\textsuperscript{+}, K\textsuperscript{+}, Ca\textsuperscript{2+}, Cl\textsuperscript{−}) and compared with experimental results obtained using flexible graphene FETs (see Fig.1). Taking into account the physiological changes of both pH and ion concentration, the scope and limitations of the intrinsic capability of graphene transistors for pH and ion sensing are discussed.

![Figure 1](image_url)

**Figure 1:** a,c: Current-voltage measurements of a flexible graphene transistor (Width: 80 µm, Length: 30µm) with \( V_{DS} = 100mV \) measured vs a Ag/AgCl reference electrode in a) 0.1M PBS solution with 0.1KCl and c) 5mM PBS solution for different pH and NaCl concentrations, respectively. b,d) CNP variations that correspond to a sensitivity of 18mV/pH and 23mV/log\(_{10}(\text{Conc})\).

References