Development and use of single-layer CVD graphene in electrolyte-gated transistors and electrochemical devices for biosensing

J. Borme1, S. Teixeira1,3, R. Queirós1, G. Machado Jr1, N. Vieira1,4, M.F. Cerqueira2, P. Alpuim1,2

1INL – International Iberian Nanotechnology Laboratory, Av. Mestre José Veiga, Braga, Portugal
2CFUM – Centre of Physics, University of Minho, Campus de Gualtar, Braga, Portugal
3College of Engineering, Swansea University, Singleton Park, Swansea, UK
4IFSC – Physics Institute of São Carlos, University of São Paulo, São Carlos-SP, Brazil
jerome.borme@inl.int

Among the many potential applications of graphene, chemical and biosensing are areas where graphene has the prospect to reach market in the short term. In transistor applications, graphene performs as a sensor due to the strong dependence of its transport properties on the charge environment [1]. In electrochemical applications, graphene provides a higher signal-to-noise level and fast electron transfer than conventional electrodes [3]. The high chemical stability of graphene with respect to saline solutions used in chemical and biological experiments also helps avoiding corrosion problems normally found with thin film devices. Low-cost commercial sensors based on screen-printed electrodes of graphene and metallic inks are widely used to benchmark graphene ability in electrochemical biosensing [2]. However, screen-printed graphene consists of randomly stacked multilayers, which precludes their use in transistors, which require the high carrier mobility found in single-layer graphene (SLG). SLG grown by CVD, on the other hand, can be used for both purposes, and provides a higher signal and of higher quality.

In this work, fabrication and operation of electrochemical sensors (ES) and electrolyte-gated graphene field-effect transistors (EGFETs) were fabricated on 200 mm oxidized silicon wafers and tested as label-free immunosensors. For EGFETs, a design was used where the conventional wire used for the gate electrode is replaced by an integrated gate coplanar to the source and drain. The contacts were processed using standard UV-optical lithography and clean-room processes. In this scheme, graphene transfer is postponed as much as possible, to avoid incompatibility with clean room processes used for patterning the other layers, while allowing for a single patterning step of graphene. The EGFETs were used as immunoassays for serpin detection. The graphene channel was functionalized using a linker (PBSE, Pyrenebutyric acid N-hydroxy-succinimide ester) followed by immobilization of anti-serpin antibody and subsequent detection of different serpin concentrations. The sensor signal will be based on the linear part of the transfer curve of the transistor (either the electron or the hole branch). The figure shows the shift in the transfer curve of the EGFET as the serpin concentration increases in the range from 0.01 ng/mL to 10 ng/mL. The sensor signal could be either the channel current for a fixed value of gate voltage or the gate voltage necessary to maintain a fixed source-drain current. In both cases, the source-drain voltage would be kept at a constant value.

A set of samples including a thin film reference electrode in platinum was also fabricated. The reference electrode is located in the same plane of the other electrodes, and close to the source and drain contacts. This supplementary electrode allows the graphene device to be operated with a potentiostat as an electrochemical device. Preliminary proof of concept showing basic operation of the sensor will be shown.

Emphasis will be given on major challenges regarding graphene sensors from the perspective of their competitiveness with existing commercial solutions in terms of fabrication and cost.
References


Figures

Figure 1: Graphene anti-serpin functionalized EGFET transfer curves for different serpin concentrations