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Control of Aryldiazonium Reactions on Graphene Field-Effect Transistors

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Introduction: Graphene is a promising nanomaterial for chemical sensors or biosensors, but functionalization of its surface is usually necessary to ensure specific interactions with the chosen analyte. Among functionalization strategies, covalent adducts are most likely to ensure stability of the functionalization during multiple flow cycles. In particular, aryldiazonium salts are known for their high reactivity with carbon allotropes. In single-walled carbon nanotubes, spontaneous covalent reactions with this reagent have been observed, notably via a strong diminution of conductance in electrical measurements and increase of D/G intensity ratio in Raman spectroscopy. However, results of this same reaction with graphene have been far less consistent, with mixed signatures of both chemisorption (covalent) and physisorption (non-covalent). This highlights the need to better understand and control aryldiazonium functionalization of graphene.

Methods and Results: Here we present novel experimental data to disambiguate the effect of aryldiazonium functionalization on graphene transport properties. We assembled an experimental setup to address an array of graphene field-effect transistors (GFETs), both simultaneously and individually. These GFETs were made of graphene grown by chemical vapor deposition (CVD), operated using a coplanar electrode in saline buffer, and functionalized with 4-carboxylbenzene diazonium tetrafluoroborate. The potential applied on the gate was varied during functionalization, which was monitored through electrical measurements and hyperspectral Raman imaging. We report a strong variation in the rate and yield of formation of covalent adducts with gate potential.

Conclusion and Significance: By incorporating past and recent experiments, we were able to explain and control graphene reactivity to the aryldiazonium chemistry with electrostatic potential. These results will be instrumental for improving the functionalization of graphene FETs with stable covalent adducts for chemical and biological sensing applications.

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