Abstract:

The goal of our project is to pattern well-defined electrodes from single-layer graphene for use in electrochemistry investigations. We have fabricated devices using both mechanically exfoliated graphene and chemical vapor deposition (CVD) graphene. Preliminary experimental data show that the graphene is chemically clean, based on measurements that the effective area of the electrodes determined by cyclic voltammetry agrees with the geometrical area. We are using these electrodes to investigate the interactions between graphene and adsorbed molecules.

Summary of Research:

The schematic design of our devices is illustrated in Figure 1. A single-atomic-layer sheet of graphene is supported on a Si-SiO₂ substrate and connected to metal leads using optical lithography. A layer of Al₂O₃ followed by a layer of parylene is deposited on top of metal leads to isolate them from the electrolyte solution to be used in the electrochemistry experiments. We pattern the parylene with an oxygen plasma etch.

We then open a window in the Al₂O₃ layer using a wet etch to expose the graphene surface. This design allows for the graphene to be the only electrically-active surface in contact with the electrolyte solution during electrochemical measurements. The size of the exposed graphene ranges from 15 × 15 µm² for mechanically exfoliated graphene to 380 × 500 µm² for CVD graphene. We use highly resistive Si wafers to reduce the capacitance of the electrode, and employ a vacuum anneal to clean contaminants from the graphene surface after fabrication. For CVD graphene devices, we are able to grow and transfer graphene sheets as large as 1.5 × 1.5 cm², so that we can form multiple electrodes on each chip and dice them into individual devices before use.

The fabricated graphene electrodes are characterized by several techniques to confirm the quality and cleanliness of the graphene. Figure 2 shows Raman spectroscopy of the mechanically exfoliated graphene and the CVD graphene. The large ratio of the amplitude of the 2D peak to the geometrical area. We are using these electrodes to investigate the interactions between graphene and adsorbed molecules.
indicate that we have high-quality single layer graphene in both types of devices. Figure 3A shows an atomic force microscope (AFM) image of a device fabricated from exfoliated graphene. The flat and dirt-free surface provides evidence for the cleanliness of the graphene after fabrication. The step height of the graphene with respect to the SiO₂ is 1 nm, in good agreement with the value for pristine graphene [1]. Figure 3B shows an AFM image of a fabricated CVD graphene electrode. In contrast to the mechanically exfoliated graphene, the CVD graphene surface has gaps and wrinkles due to the transfer of CVD graphene from the Cu substrate onto the Si wafer. The particles shown in the image are Cu and FeCl₃ introduced during the transfer process. Despite the fact that both Cu and FeCl₃ are electrochemically active; we do not see any signals from these materials in our cyclic voltammogram measurements. We conclude that particles are either underneath the graphene or wrapped by graphene and so do not interact with the solution during measurement.

We are using the fabricated graphene electrodes for electrochemical measurements. Figure 4 shows the measurement setup. A graphene electrode, a Ag/AgCl reference electrode and a Pt counter electrode are connected to a CHI 900 potentiostat in three-electrode configuration. Cyclic voltammograms are measured with both mechanically exfoliated and CVD graphene. The effective area of the electrodes can be determined from the cyclic voltammogram measurements. For both exfoliated and CVD graphene the effective electrochemical areas are equal to the geometric areas within experimental accuracy (about 20%). We are currently using electrochemical techniques to characterize the adsorption of molecules on the graphene and charge transfer between these adsorbed molecules and graphene.

References: