



Reversible oxidation of graphene

A step towards better performance graphene-based electronics

The collaborative works between US and Japanese researchers have developed a new way to oxidize graphene reversibly that could be a step closer to the creation of high performance graphene-based electronics.

<Introduction>

Graphene, a one-atom thick, honeycomb-shaped lattice of carbon atoms with exceptional physical and electronic properties, is very promising for application in next-generation electronics. Many experts consider graphene as the rival to silicon in creating faster, thinner and flexible electronic devices. But major challenge in realization of graphene-based electronics is to achieve the tunable electronic properties of graphene. Unlike semiconductor such as silicon, pure graphene is a zero band-gap material, making it difficult to electrically `turn-off` the flow of current through it. Therefore, pristine graphene is not appropriate for digital circuitry that comprise vast majority of integrated circuit.

In an effort to control its properties and make the graphene more functional, researchers are investigating methods for chemically altering the graphene. The most widely used strategy is the "Hummers method" a process developed in 1940s that oxidizes graphene. However, the chemical inhomogeneity and irreversibility of the resulting graphene oxide surface arise because of using aggressive oxidizing agents. This collaborative works between US and Japanese researchers of different institutes including Gunma University, The University of Tokyo, RIKEN and KEK have recently developed a new method to oxidize graphene that overcomes the collateral damage encountered in Hummer's method.

<Results and Discussion>

Performing chemical reactions on graphene is very difficult because of the bonding nature of the carbon atoms forming hexagonal networks. Typically researchers employ strong acidic conditions, such as those utilized in the Hummer's method, that damage the honeycomb-shaped lattice and result in a material that is difficult to control. In our method, we leaked oxygen gas (O_2) into an ultra-high vacuum chamber, which housed a hot tungsten filament (~ 1500 °C) that causes the oxygen molecule to dissociate into atomic oxygen. The highly reactive oxygen atoms then uniformly bonded to the graphene lattice.

The graphene oxide formed in ultra-high vacuum is chemically homogeneous and thermally reversible, which can lead to well-controlled properties and be exploited in high performance electronics. The chemical homogeneity and reversibility of the oxidized graphene are characterized by various surface probe techniques including scanning tunneling microscope (STM) and x-ray photoelectron spectroscopy (XPS). A typical example of STM images of pristine and oxidized graphene on silicon carbide (SiC) substrate is shown in figure 1. Figure 2 shows a series of C 1s XPS spectra of pristine graphene on a SiC substrate (black), oxidized graphene by atomic oxygen in UHV (red) and annealed graphene at 260 °C (blue).

We think it is a step in the right direction to explore the means of chemically modifying graphene to develop a wide variety of materials, similarly as scientists did for plastics in the last century.



Figure 1. Scanning tunneling microscope (STM) images of before (left) and after (right) oxidation of graphene on silicon carbide (SiC) substrate. Pyramidal-shaped protrusions in the right image are the individual oxygen atom bonded to graphene



Figure 2. C1s X-ray photoelectron spectra measured at KEK-PF BL13A (2009S2-007). Pristine epitaxial graphene on a SiC substrate (black). Oxidized graphene using atomic oxygen in UHV (red). The clean graphene was recovered after heating at 260 °C (blue).

<Publication>

Md. Zakir Hossain, James E. Johns, Kirk H. Bevan, Hunter J. Karmel, Yu Teng Liang, Shinya Yoshimoto, Kozo Mukai, Tatanori Koitaya, Jun Yoshinobu, Maki Kawai, Amanda M. Lear, Larry L. Kesmodel, Steven L. Tait, and Mark C. Hersam, "Chemically homogeneous and thermally reversible oxidation of epitaxial graphene" *Nature Chem.* 4, 305-309, **2012**.

<Contacts>

Jun YOSHINOBU, Professor, The Institute for Solid State Physics Phone: +81-4-7136-3320 Fax: +81-4-7136-3473 E-mail: yoshinobu@issp.u-tokyo.ac.jp

Md. Zakir Hossain, Research associate, Gunma University http://asrldu.dept.med.gunma-u.ac.jp/hossain/Md.Zakir_Hossain.html E-mail: zakir@gunma-u.ac.jp