Transferring Chemical Vapor Deposition Grown Graphene

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Abstract:

Since the unique properties of graphene have made it a candidate to be integrated into a wealth of applications, an industrial process for creating graphene should be developed. Although the technique to produce graphene on copper foils using chemical vapor deposition (CVD) [1] has been discovered by the Ruoff Group, it is still needed to develop an efficient way to transfer graphene onto arbitrary substrates, while maintaining its electrical and mechanical properties over large length scales. Our research investigated methods to optimize graphene transfer using two different 'carrier' materials. First, we explored using poly(methyl methacrylate) (PMMA) as a carrier for the graphene and tried to optimize the quality of the transferred graphene. We also looked into the use of thermal release tape as a carrier and investigated the role that adhesion plays in transfer.

Experimental Process:

By flowing hydrogen and methane through a high temperature furnace, monolayer graphene can be formed on the surface of the copper foils. The methane is catalytically decomposed on the copper surface at the high temperature (typically around 1000°C), which leads to nucleation, island growth, and finally complete coverage with a poly-domain film of monolayer graphene (Professor Ruoff and colleagues use 'domain' rather than 'grain,' as a grain is typically considered to be three dimensional). Because a copper foil cannot be used as a substrate in the electrical testing of graphene, the graphene must be transferred to a different substrate for various applications.

Our transfer process started with etching one side of the copper using an iron nitrate solution [2]. After a short copper etch, we applied a solid carrier to help transfer the graphene; subsequently, the rest of the copper was etched away. The carrier with graphene was then transferred onto a substrate like silicin dioxide / silicon (SiO_2/Si). After removal of the carrier, only graphene was left on the substrate. This transfer procedure can be modified with the objective of getting the highest quality graphene after transfer.

The first portion of our research focused on using PMMA from Aldrich as a graphene carrier [3]. Our first experiment was to explore the method in which graphene contacted the substrate, in our case, SiO_2. After the PMMA layer was dried, and the copper foil was fully etched, the graphene/PMMA stack was placed on top of a silicon wafer with graphene contacting the SiO_2. The first method of creating contact was to apply an additional drop of liquid PMMA. This method, invented by the Ruoff group, allowed the existing dried PMMA to dissolve, and the graphene to relax onto the substrate.

The next method to create contact of graphene to substrate was to heat the silicon wafer with the graphene/PMMA stack on top. We learned that heating the wafer allowed PMMA to become malleable, and our goal was to see if the graphene could thereby relax on the wafer surface.

We compared the results of each method using an optical microscope. Generally, transfers where the Si wafer substrate had been heated had fewer voids in the graphene film compared to transfers that used additional liquid PMMA. Decreasing the number of voids is important when configuring graphene in electrical and mechanical experiments.

In addition to exploring contact methods using PMMA, the removal of the PMMA was also investigated. The first method tried was to try to wash away the PMMA using acetone. Another method tried was to evaporate the PMMA using a furnace at 400°C with H_2 and N_2 gases. Comparing the results of each removal method with optical microscopy, it seems that evaporation removed PMMA to a greater extent than the acetone wash. Unfortunately, the evaporation process did create amorphous carbon, verified using Raman spectroscopy.

Thermal release tape (TRT) from Nitto Denko was also tested as a solid graphene carrier [4]. TRT was applied to graphene on copper foils and the TRT/graphene was transferred to a target substrate after dissolving copper. When the substrate was heated to a specific temperature, TRT released the graphene onto the target substrate.

We investigated three different types of TRT for graphene transfer. Figures 1-3 show optical microscope images of TRT transfers using TRT with increasing adhesion strengths. Our findings suggest that the strongest TRT used yielded the least amount of voids after transfer, but contained the most surface residue on graphene.

Results and Discussion:

In summary, after comparing methods for creating contact using PMMA, we found that the number of voids can be decreased by heating the substrate instead of using additional liquid PMMA. We found that evaporating PMMA, rather than using acetone to try to remove it by dissolution, can decrease the amount of residual PMMA left on the graphene surface. Our research also suggests that the strength of the adhesion of the TRT is apparently important in the transfer of graphene in terms of the number of voids per unit area in the graphene after transfer.

Future Work:

Our research efforts suggest directions for further work, and more measurements such as sheet resistance and transmittance should be conducted in order to further test which methods optimize transfer.

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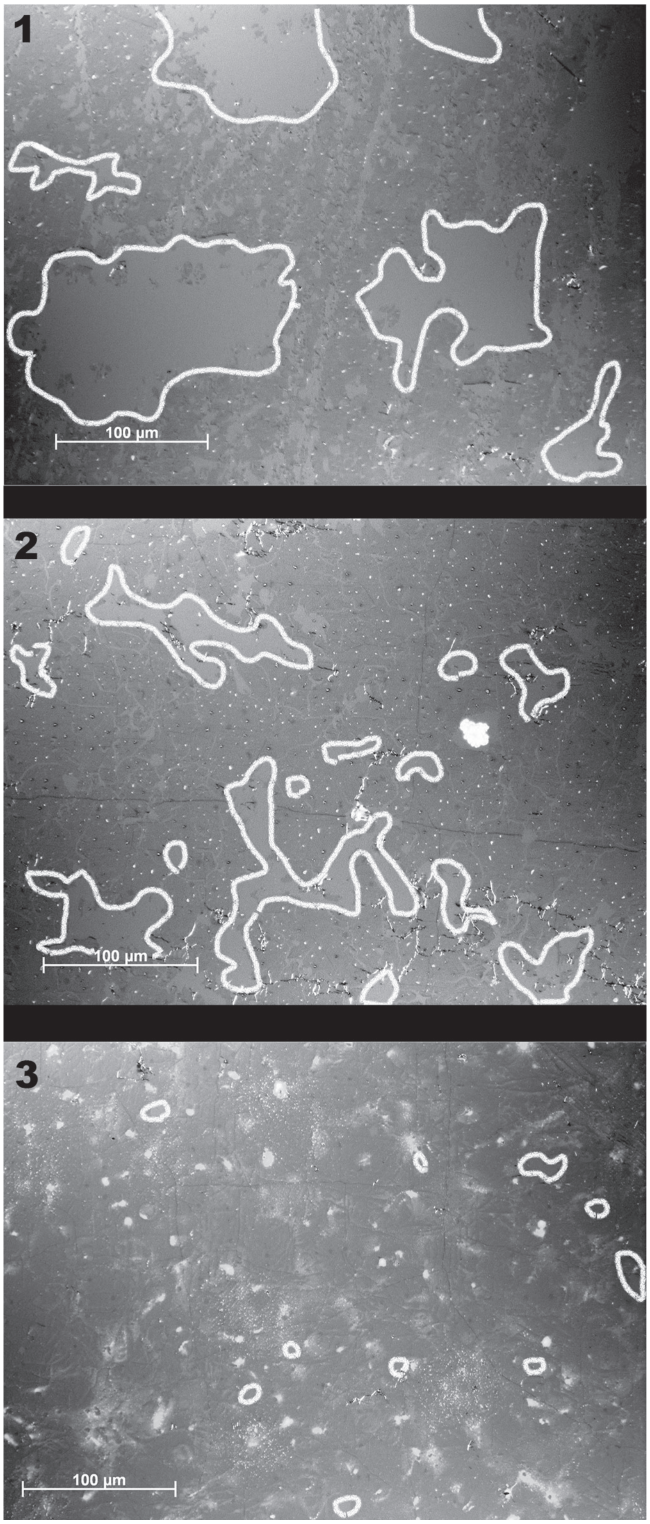

Figures 1-3 show graphene transfers using thermal release tape. As adhesion strength increases, the amount of voids decreases.