Growth of CVD-graphene on thermally annealed and electropolished Cu substrates

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The growth of graphene has been demonstrated using chemical vapor deposition (CVD) on high purity polycrystalline (99.99% purity) and single crystal Cu foils. Polycrystalline Cu foils are much more attractive than single crystal Cu due to extensive availability and lower costs. However, Cu foils (~25 µm) used for graphene growth are ridden with surface defects such as native oxide of Cu (Cu(I)O, Cu(II)O), high root mean square (RMS) surface roughness (> 100 nm) and non-equiaxed grain structures (~ 1-10 µm) with elongated grain boundary due to its polycrystallinity. These defects arise mainly because the Cu foils are prepared using metallurgical cold rolling which imprints contaminants such as Al and induces non-equiaxed Cu grains. Due to the highly unsuitable nature of the Cu foils as obtained from vendors such as Alfa Aesar, it is challenging to obtain controllable, and reproducible large area graphene growth. To mitigate the unsuitable Cu surface, research groups have employed a variety of surface preparation techniques prior to graphene growth [1]: by removing the Cu native oxide using thermal annealing in H_2 environment or dipping in Cu foils in nitric acid and using chemical mechanical polishing to reduce the surface roughness. These techniques offer adequate improvement of the Cu surface by removing native oxide and reducing the surface roughness. But these techniques still do not achieve nanometric level RMS roughness (~1-5 nm) desired for continuous large area graphene growth. Simultaneously, employing these techniques also introduce new type of surface defects on Cu in the form of etch pits due to the use of corrosive acids such as nitric acid for surface preparation of Cu prior growth.

In our previous work on hexagonal boron nitride (h-BN) [2], we used a two-step thermal annealing (in H_2 environment at 1030°C) and electropolishing procedure to make Cu surface much more suitable for growth by removing the native oxide, and reducing the surface roughness. When we use this two-step procedure, we consistently observe a larger Cu grain size (> 100 µm) measured using optical and scanning electron microscope (SEM) micrographs, and surface roughness at the nanometric level (1.1-1.4 nm) as measured on a 100 μm^2 scan area using atomic force microscopy (AFM). Growth of h-BN on these Cu foils lead to larger (~250%) and fewer (~60%) h-BN crystals as compared to growth on only thermally annealed Cu. We use the same surface preparation conditions to assess if this technique can be extended to graphene which is crystallographically similar to h-BN. We will report the details of our two-step surface preparation process and also the graphene growth metrics on these thermally annealed, electropolished Cu foils. SEM, AFM and Raman spectroscopy will be used to assess the size, count, thickness and quality of the graphene crystals grown.

References:

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