

# Supporting Information For:

## Anisotropic Etching and Nanoribbon Formation in Single-Layer Graphene

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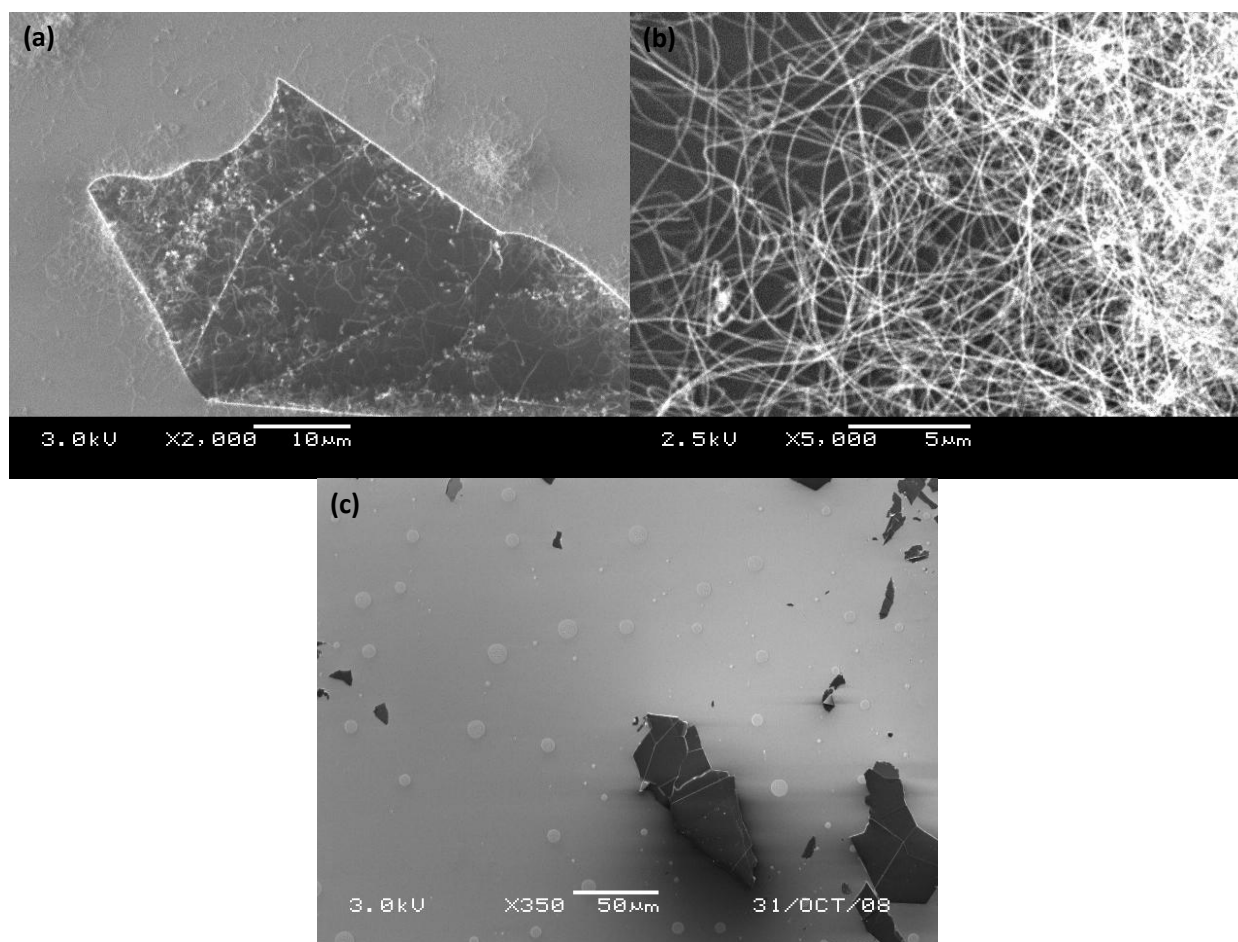
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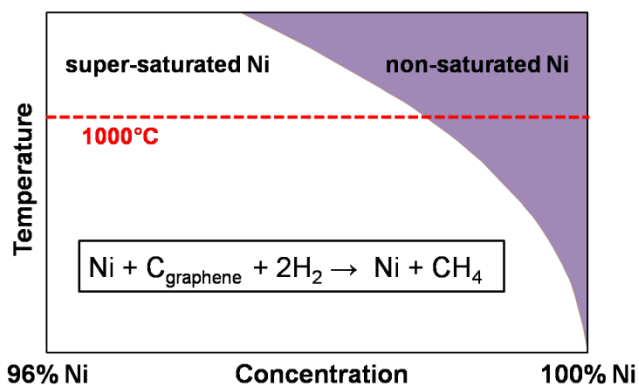


**Figure S1. SEM images of graphite after etching process (a) Carbon nanotubes grow on a sample with a low concentration of applied  $\text{NiCl}_2$  solution (b) Higher magnification of a cluster of CNTs. (c) A higher concentration of  $\text{NiCl}_2$  results in the formation of Ni islands and no carbon nanotubes are formed.**

One of the main obstacles in finding the ideal conditions for producing cuts in single layer graphene is the growth of carbon nanotubes (Figure S1a). Although the growth of carbon nanotubes and the production of cuts are not mutually exclusive processes (images containing both CNTs and cuts have been measured), cuts produced along with CNTs are generally not optimal and it is very difficult to image the cuts in the presence of CNTs. As mentioned in the main text, the catalytic hydrogenation of graphene is the effective reverse process for the catalytic growth of carbon nanotubes<sup>1, 2</sup>. Depending on the local concentrations of the respective reactants, the results can change drastically.

The first obvious culprit responsible for the production of CNTs is organic contaminants remaining on the sample surface during the etching process. This is inferred from the observation of CNTs growing predominantly from residue left behind from tweezer handling, or from tape adhesive residue from the mechanical exfoliation process. These organic contaminants provide the carbon source for the production of carbon nanotubes. This is highlighted by the fact that even when performing the etching process on just a SiO<sub>2</sub> substrate, without any exfoliated graphene, it was found that it was possible to grow CNTs without addition of any extra hydrocarbons except the contaminants on the surface. This underscores the need for special cleaning during sample preparation, most importantly to remove adhesive residue after the exfoliation process by heat cleaning under forming gas (500°C for 15min under Ar:H<sub>2</sub> flow (850:150 sccm)).

Even with organic contaminants minimized, it is still possible to grow CNTs since the graphite itself acts as a carbon source<sup>3</sup>. The etching process releases the graphite's carbon as methane which under certain conditions is sufficient to cause CNT grow. These conditions are determined from the concentration of interstitial carbon within the Ni nanoparticles, as described by the binary phase diagram of Nickel and Carbon (Figure S2). If the concentration of carbon in the Ni nanoparticles reaches supersaturation, the nanoparticle can begin to expel carbon nanotubes<sup>1, 2</sup>. We find that lowering the concentration of Ni nanoparticles (by applying NiCl<sub>2</sub> solution with concentration < 0.05mg/mL) typically causes the growth of CNTs, but higher concentrations of nanoparticles (NiCl<sub>2</sub> solution with concentration > 2.6 mg/mL) result in no CNTs. It appears that at low concentrations of Ni, the nanoparticles more easily reach the supersaturated state which is conducive to CNT growth. At high concentrations of Ni, we suggest that large aggregation of Ni "islands" on the substrate surface (Figure 1b) absorb enough excess methane such that supersaturation is never reached.



**Figure S2. Binary Phase Diagram of Nickel and Carbon<sup>4</sup>.** In the non-saturated region (purple) fcc Ni stably coexists with interstitial carbon atoms. In the super-saturated region (white) the carbon is no longer stable within the Ni and will be expelled. (Inset) Summary of the hydrogenation reaction which drives the etching process.

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