Ultrathin metal films for direct thermochemical vapor deposition on dielectric substrates of single and a few layer graphene

Miriam Marchena, Valerio Pruneri

ICFO - Institut de Ciències Fotòniques - Parc Mediterrani de la Tecnologia, Av. del Canal Olímpic s/n 08860 Castelldefels, (Barcelona), Spain
ICREA - Institució Catalana de Recerca i Estudis Avançats, 08010 Barcelona, Spain
miriam.marchena@icfo.es

Abstract

The thermochemical vapor deposition of single-few layer graphene on Ni and Cu metal films (UTMF) on glass substrates was proposed. In this paper, we investigate the possibility to use thinner metal films than previous studies, so called Ultrathin Metal Films (UTMFs), as thin as 7nm, as catalyst. We show that one can obtain improved quality graphene over a larger area than previously demonstrated. The technique allows direct growth of graphene without significant increase of light reflection and absorption, thus offering the possibility to avoid cumbersome transfer after standard growth on much thicker (25-35 μm) metal foils.

After evaluating the effect of the different process parameters, Ni of 7 and 50 nm thickness was found to be more effective metal catalysts for graphene growth than Cu, despite the fact that solubility in Ni is higher and for this reason the tendency of multilayer graphene formation is increased. Ni is less prone to dewetting effects, i.e. metal retraction due to substrate heating. Experimentally, the minimum temperature at which graphene starts depositing has been determined to be 700ºC, and two different mechanisms of growth have been observed depending on the temperature and time: a) growth of graphene on top of ultrathin Ni without dewetting, at 700ºC, by increasing the reaction time, and b) growth of graphene in the dewetted area of Ni by increasing the temperature to 900ºC.

References

Figures

Table 1. Summary of the process conditions and characterization results

<table>
<thead>
<tr>
<th>Metal Catalyst</th>
<th>Sample</th>
<th>Thick (nm)</th>
<th>T (ºC)</th>
<th>P (mbar)</th>
<th>t (min)</th>
<th>CH₄/H₂</th>
<th>I₂D/I₂G</th>
<th>I₀/I₂G</th>
<th>FWHM 2D (cm⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni</td>
<td>S1'</td>
<td>5/7/50</td>
<td>1000</td>
<td>7</td>
<td>30</td>
<td>1.5</td>
<td>0.297/1.331/2.334</td>
<td>1.394/0.865/0.047</td>
<td>43.3/35.9/26.4</td>
</tr>
<tr>
<td></td>
<td>S2'</td>
<td>50</td>
<td>700</td>
<td>7</td>
<td>30</td>
<td>1.5</td>
<td>5.847</td>
<td>1.118</td>
<td>22.8</td>
</tr>
<tr>
<td></td>
<td>S3'</td>
<td>50</td>
<td>900</td>
<td>7</td>
<td>30</td>
<td>1.5</td>
<td>2.280</td>
<td>0.492</td>
<td>29.7</td>
</tr>
<tr>
<td></td>
<td>S4'</td>
<td>7/50</td>
<td>1000</td>
<td>7</td>
<td>15</td>
<td>1.5</td>
<td>0.350/1.789</td>
<td>0.904/0.911</td>
<td>40.5/36.4</td>
</tr>
<tr>
<td></td>
<td>S5'</td>
<td>7/50</td>
<td>1000</td>
<td>7</td>
<td>60</td>
<td>1.5</td>
<td>0.425/2.793</td>
<td>1.250/0.528</td>
<td>34.4/27.6</td>
</tr>
</tbody>
</table>
Figure 2. Raman microscope pictures after graphene CVD on Ni (50 nm thick) at: a) S2’ conditions. Starting of Ni dewetting where graphene grows suspended. Inset: SEM of the dewetted Ni, b) S3’ conditions. For increasing temperature, larger areas are dewetted and graphene progressively deposits in those areas.

Figure 2. Raman analysis after graphene CVD on Ni UTMF: a) Dependence on deposition temperature, b) Dependence on deposition time and c) Dependence on Ni thickness.

Figure 3. SEM characterization of CVD graphene on Ni UTMF: a) S1’Ni50nm. Surface is totally dewetted and with high roughness, b) S2’Ni50nm at 700°C. Arrows indicate where graphene is suspended between the dewetted UTMF of Ni, c) S3’Ni50nm at 900°C. Larger area has been dewetted where graphene grows.