Pre-Patterned CVD Graphene: Influence of ALD deposition parameters on $\text{Al}_2\text{O}_3$ and graphene layers

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Abstract
Fabrication of graphene nanostructures is of importance for both investigating their intrinsic physical properties and applying them into various functional devices. Our group has been developing studies on direct synthesis of graphene from an area-selectively passivated catalyst substrate in order to generate patterned graphene of high quality. Here we present a further study on this subject focusing on ALD deposition of $\text{Al}_2\text{O}_3$ to produce patterned graphene through area-selective CVD growth. We present the influence of number of cycles and purging time on $\text{Al}_2\text{O}_3$ deposition uniformity and graphene quality. The deposited $\text{Al}_2\text{O}_3$ films and the graphene layers are investigated in terms of morphology and electrical properties. It was obtained an optimized condition, in which it was possible to prepare uniform $\text{Al}_2\text{O}_3$ layers with negligible mobility, and defect-free graphene with sheet resistance around 320 ohm/square.

Introduction
Pre-patterned graphene could potentially produce higher quality graphene devices since the patterning step is carried out before the graphene is synthesized and the risk of contamination is minimized [1,2]. We here use ALD to generate patterned graphene through area-selective CVD growth. The method relies on the passivation of defined areas of copper foil and the subsequent selective growth of graphene in the unpassivated regions. Our group already developed works on this subject, in order to obtain high quality patterned graphene with low cost and high resolution. Hoffman et al reported the possibility of barrier guided CVD generates high quality graphene on pre-patterned substrates, and thus open up the possibility to efficiently produce interconnects for a wide application [3]. Thus, this present work focuses on a further study on pre-patterned graphene, centering on the influence of ALD deposition parameters on graphene and passivation layers quality.

Experimental Procedure
In order to protect selected areas of the copper foil during $\text{Al}_2\text{O}_3$ deposition was used a PMMA mask. The pattern was draw using a shadow mask and a brush. The deposition of $\text{Al}_2\text{O}_3$ was carried out using a homebuilt ALD system. In each cycle trimethylaluminum and water were released for 15ms. As a purge gas was used nitrogen and the temperature during the deposition was 150°C. It was used 120 and 200 cycles and 30 sec and 45 sec of purging time. After the deposition the PMMA mask was dissolved in acetone with sonication for 2 hours. Graphene growth was carried out in a LPCVD furnace at 1000 °C for 30 minutes flowing 50 sccm of hydrogen and 20 sccm of methane. Graphene transfer was carried out using wet transfer technique.

Results and Discussion
Increasing the purging time from 30 seconds to 45 seconds it seems to influence both graphene and $\text{Al}_2\text{O}_3$ quality. It is known that increasing the purging time in the same number of cycles it is possible to remove non-reacted reactants much more efficiently. AFM phase images of graphene showed that the graphene grown after the deposition with 30 seconds of purge contain more impurities than the graphene grown after the deposition with 45 seconds. The PMMA mask did not protect the cooper foil efficiently when more reactants are present in the chamber. It was observed the presence of two phases on 1(a) while on 1(d) we can observe a smooth graphene surface free of impurities. Increasing the number of cycles and purging time it was possible to obtain better uniformity of $\text{Al}_2\text{O}_3$ layers, the best result was achieved with 200 cycles and 45 sec of purge, showed by optical microscopy in figure 2.

Comparing the sheet resistance of the graphene layers and the passivation layer in which condition, it was observed that the best result for the graphene layer was 328 ohm/square while the worst was found around 2927 ohm/square. Regarding the sheet resistance of the passivation layer all the conditions presented values greater than 1000 ohm/square, with electrical mobility ranging between 19-60 (cm$^2$/Vs), but on the deposition condition of 120cycles/45 seconds which was not found any mobility. This behavior can allow the direct integration of resistive elements in graphene circuits during growth, since the passivation layer is isolating graphene areas from one another.
Conclusion
In this work the influence of parameters of ALD deposition of Al₂O₃ on pre-patterned graphene was reported. The best results were obtained using 200 cycles and 45 seconds of purge, with an uniform Al₂O₃ layer with negligible mobility, and defect-free graphene with sheet resistance around 320 ohm/square.

References

Figures

Figure 1: AFM images of graphene layer grown after the Al₂O₃ deposition with: (a) 120 cycles with 30 seconds purge, (b) 120 cycles with 45 seconds of purge, (c) 200 cycles with 30 seconds purge, (d) 200 cycles with 45 seconds purge.

Figure 2: Optical images of Al₂O₃ layers deposited with different conditions (a) 120 cycles with 30 seconds of purge, (b) 120 cycles with 45 seconds of purge, (c) 200 cycles with 30 seconds of purge, (d) 200 cycles with 45 seconds of purge.

Figure 3. Electrical properties of graphene and Al₂O₃ passivation layer

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<tr>
<th>Al₂O₃ layer</th>
<th>Mobility (cm²/(V s))</th>
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<tbody>
<tr>
<td>120/30</td>
<td>19</td>
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<tr>
<td>120/45</td>
<td>-</td>
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<td>200/30</td>
<td>60</td>
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