Growth of Single-Crystal Bi-Layer Graphene Using Alcohol CVD

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Graphene, an sp²-bonded single-layer carbon sheet, is an attractive material due to its extraordinary mechanical, thermal, and electrical properties. However, single-layer graphene (SLG) is the zero band-gap material and unsuitable for some applications which need finite energy gaps. Several groups reported that a bandgap up to 250 meV can be opened by an external electric field in AB-stacked bi-layer graphene (BLG) [1]. Therefore, efficient synthesis of BLG is essential for electronic applications of graphene. Chemical vapor deposition (CVD) is a reliable method for the synthesis of large-scale graphene. Since grain boundaries in polycrystal graphene degrade its superior properties, many efforts have been focused on enlarging the size of single-crystal graphene. As a result, single-crystal SLG of centimeter sizes has been synthesized using CVD, but the size of single-crystal BLG domains is still limited.

In this study, we investigated the method for the single-crystal BLG synthesis and succeeded in synthesizing BLG domains of several hundreds of micrometers. Graphene was synthesized using alcohol catalytic chemical vapor deposition (ACCVD) [2]. We used a copper foil as the catalyst and oxidized and folded it into a pocket before synthesis. CVD was performed at 1065 °C with various pressures of Ar/H₂ and ethanol. We found that SLG, BLG, and multi-layer graphene can be grown by controlling the total pressure of the CVD condition. Figure 1 shows large-size single-crystal BLG together with SLG. Figure 2 is the relationship between the graphene size and the growth time. This revealed that there is an incubation time for BLG growth after starting alcohol supply.





Fig. 1 Optical image of SLG and BLG.

Fig. 2 Relationship between BLG size and growth time.

[1] Y. Zhang *et al.* Nature **459**, 820 (2009).
[2] X. Chen *et al.* Carbon, (2016) in press.
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