Temperature dependence of twisted graphene formation on graphene/silicon carbide template

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[Introduction] Twisted few layer graphene (FLG) has recently attracted great attention due to the appearance of the exotic electrical properties, such as superconductivity and Mott-like insulator [1]. In previous study [2], we reported that the FLG was synthesized by overlayer growth of graphene on a monolayer graphene template using a chemical vapor deposition (CVD) method [3]. We found that the grown 2D graphene islands have the random twisted angles due to the coalescence of islands, leading the formation of the weak interlayer coupling like a turbostratic stacking. In this study, to understand the growth mechanism of twisted FLG, we examined the temperature dependence systematically and proposed the vertical and lateral morphology model in graphene islands grown on graphene template.

[Experiments] The 2D graphene islands with a monolayer step height were grown on silicon carbide with continuously oriented monolayer graphene via CVD with ethanol as a carbon feedstock. The growth was carried out in an infrared heating furnace at the varied temperature from 900ºC to 1450ºC. The atomic force and scanning tunneling microscopes (AFM/STM) were used to evaluate the surface morphology.

[Results and discussion] In terms of temperature dependence, Fig. 1 shows the distribution of layer distance and the approximate average size of each island, which are statistically obtained from AFM images as shown on the upper right. The layer distances indicated in the histogram are defined by the cross-sectional analysis from AFM images, and “0 nm” means the islands not reaching one layer (about 0.35nm). The peak value of the distribution at each temperature is marked. At relatively lower temperature, the counts of isolate islands distribute to longer layer distance and the average radius was preserved to be smaller clusters. At relatively higher temperature, exceeding 1300ºC as critical temperature, the graphene islands form after nucleation and tend to stack into multilayer graphene as the temperature increases. Additionally, while the average radius of higher temperature goes larger, the number of graphene islands reduces due to the combination of small islands. It is verified by STM analysis with different moiré pattern that the large pieces of the graphene islands are formed by coalescence among the small islands via the lateral growth mode, as the growth model indicated schematically in Fig. 2. These results demonstrate that the deposition of carbon atoms changes from non-planar cluster formation to coalescent graphene islands at the critical temperature around 1300ºC.

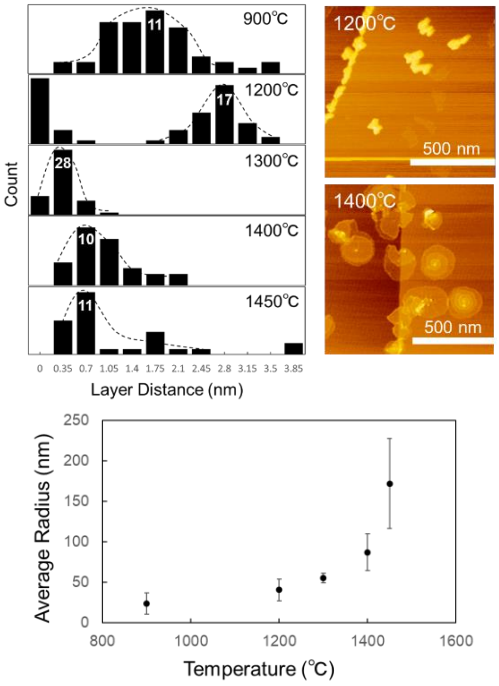

Fig. 1 Statistical data from AFM images with vertical and lateral morphology in graphene islands

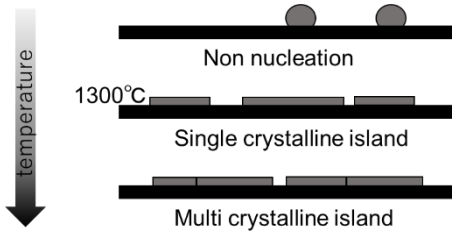

Fig. 2 The sketch of the tendency to crystallize with the temperature increasing.

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