INITIAL GROWTH OF GRAPHENE ON Cu SUBSTRATE

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Graphene has been the hottest topics in condensed matter physics and materials ever since the mechanical expoliation of a single layer from the bulk graphite in 2004[1]. It gives a sudden impact of the realization of Dirac fermions on table top followed by many extremal characteristics including room-temperature quantum Hall effect, high mobility and Klein tunneling etc. Followed by this mechanical expoliation of a single layer graphene, epitaxial graphene with the SiC substrate was believed to give easy way in UHV to characterize its electronic structures with an angle-resolved photoemission spectroscopy[2]. However, these two methods have the problems towards the electronic application. With the full experience of metal catalyst in CNT production, people used the metal substrates such as Ni, Cu, Pt, Ru for the formation of graphene on these substrates[3]. However, the solubility of carbon in these metals divides the process into two categories to form a graphene. Low solubility metal substrate such as Cu shows pure CVD process to form the graphene while high solubility metal substrate such as Ni uses the surface segregation upon rapid cooling. For the application of this graphene to electronic device, the information on the grain boundary is quite essential. The atomic structure of the grain boundary in CVD grown graphene has been recently reported[4] and it forms a pentagon or heptagon. To minimize the effect of this grain boundary in electronic device, one needs to grow larger domain sized graphene. However, the precise understanding of the CVD process is not complete so far. In this report, we show the precise control of all the parameters in CVD process in graphene growth and present the way to form larger grain in CVD growth. To change this gapless graphene to form a gap, it has been theoretically predicted that biasing or cutting it in nano-scale is necessary. We will add one simple way to form the graphene nano-ribbon(GNR) using the electron beam technique with this CVD grown graphene.

Experiments have been done at the ALD chamber to control the precise ratio of gas mixture and temperature of the substrate. This system uses the halogen lamp instead of filament heater used in conventional CVD system, so that the rapid heating and cooling is possible. This is quite essential since the growth time for a single layer of graphene is few tens of seconds so that the initial growth condition can be checked with the time interval of few seconds. The graphene was synthesized on the polycrystalline Cu foil. Cu foils were heated under H_2 gas pressure (1 sccm) up to 990 ¡C for 30 min. CH_4 and H_2 mixture gases were simultaneously flowed into the chamber for a desired growth time. Then the temperature of the furnace was immediately lowered to RT with cooling rate $\approx$ 40 ¡C/min. The graphene samples were analyzed by SEM.

To optimize the process of the graphene growth, the ratio of gas mixtures(H_2 and CH_4), the flow rate of mixture gas and the growth temperature are the parameters to control the growth. Fig. 1 shows the typical examples of two different growth modes depending on the temperature and the carbon contents on the surface. The fractal growth shows fairly large grain size but it is not easily found at high temperature growth. The island growth is dominant at high temperature, but the grain size cannot exceed that of the fractal growth. If the growth temperature is low, then there were more nucleation sites, so that the grain size becomes low. We needs to control the flow of mixture gas to obtain the fractal growth initially at high temperature. High temperature annealing(or growth) is the essential step for the defect-free graphene growth. CH_4, which has the closed-shell covalent structure, has very low sticking coefficient with metal substrate. The graphene is formed well above 1100 K by the repeated thermal decomposition of CH_4 to higher hydrocarbons. We can use this for the formation of graphene nano-ribbon using electron beam technique. We will show fairly regular pattern of graphene nano-ribbon.

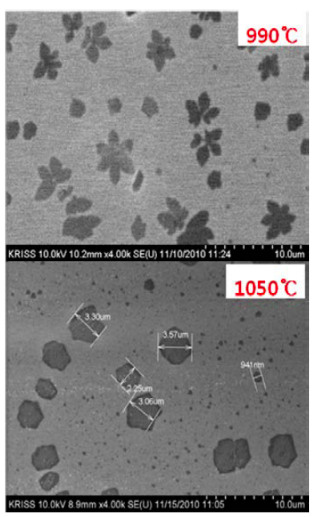

Figure 1: With different gas mixture ratio between (H_2 and CH_4), by controlling the growth temperature, initial growth pattern can be polygons or fractals.

In conclusion, we show to control the growth of graphene, especially the grain size. Also nano-meter scale graphene nano-ribbon can be easily made with the use of electron beam technique combined with the thermal decomposition process control.

References

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