Vertical and Longitudinal Multi-layer Graphene Growth Using a New Heat-Beam assisted CVD

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ABSTRACT

A vertical and longitudinal multi-layer graphene sheet has been grown on the substrate by a catalytic Chemical Vapor Deposition (CVD). For the CVD, we employed a new annealing method called "Heat-Beam (HB)", in which high temperature inert gas was blown directly to the chip surface in an inert gas-flow chamber, and a safe and chemically stable gas, was used as a hydrocarbon gas source. Although the possible formation mechanism of such an interesting graphene structure has not yet been made clear, their electrical, thermal and mechanical properties should be very promising from the point of view of electrical applications. The advantages of HB-CVD include not only a steep temperature distribution in depth, but also better scalability for mass production systems.

Background & Objectives

Graphene formation methods

- Chemical vapor deposition (CVD) [1]
- Epitaxial growth on a SiC [2]
- Reduction of graphite oxide [3]
- Laser annealing method [4]
- Annealing of amorphous carbon [5]

Many issues to be solved for mass production, such as cost, throughput, and quality.

One of the possible solutions must be **Ambient Pressure Continuous CVD with conveyor belt** [6]

However, there are still some issues: conventional heating methods (~800℃)
- ×leads to apparatus distortion
- ×limit applicable substrates with high heatproof

New Heat-Beam (HB) CVD

-No vacuum chamber
⇒Low cost and High throughput
-No apparatus distortion
⇒Large-scale mass production machine
-Potential for roll-to-roll system
⇒High throughput

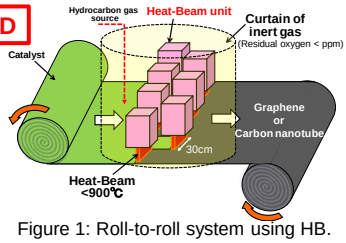
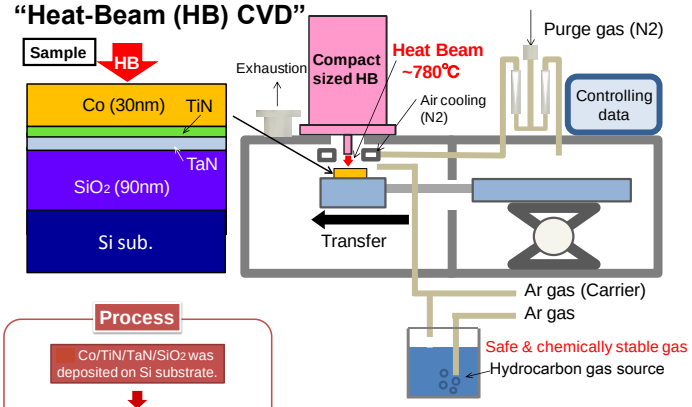


Figure 1: Roll-to-roll system using HB.

Experiments

"Heat-Beam (HB) CVD"



Process

1. Co/TiN/TaN/SiO_2 was deposited on Si substrate.
2. Filling the chamber with N_2. (Residual oxygen in the chamber<ppb.)
3. Hydrocarbon gas and Ar gas are added into the chamber.
4. Heated N_2 gas is blown down to the substrate.

Figure 2: Heat-Beam CVD machine.

【Other conditions】
- Catalyst: Co (30nm thick)
- Purge gas or inert gas curtain: N_2
- A new Hydrocarbon gas source
- Growth temperature: **~650ºC**
- **Ambient pressure**

Results and Discussion

"Raman spectrum"
- G/D ratio : 2.5
- Peak of cobalt oxide.

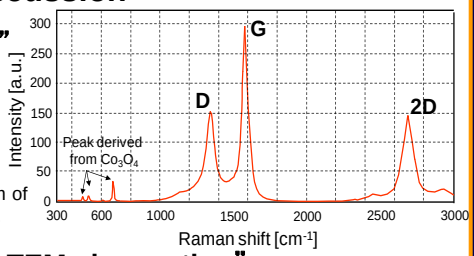


Figure 3: Raman spectrum of HB-CVD grown graphene.

"Cross-sectional TEM observation"

Multi-layer graphene (more than a few hundred layers) was successfully grown by HB-CVD for the first time. The Inter-layer distance is about 0.35 nm. Both vertical and longitudinal multi-layer graphene (Fig.4(a)) and horizontal multi-layer graphene structures (Fig.4(b)) were observed by cross-sectional TEM observation.

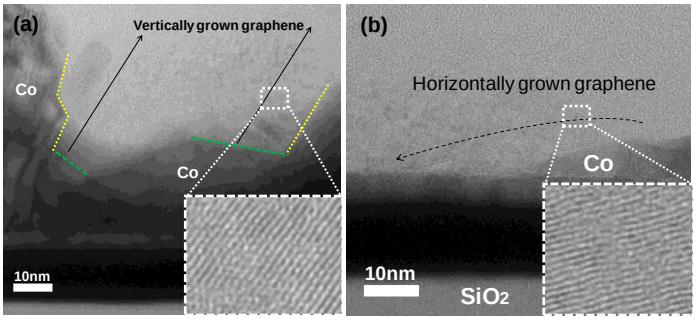


Figure 4: Cross-sectional TEM images and models of graphene growth.

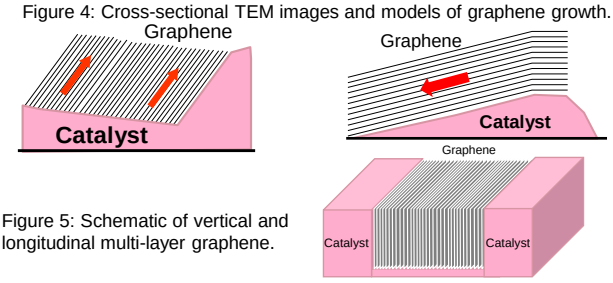


Figure 5: Schematic of vertical and longitudinal multi-layer graphene.

Conclusions

A new structure of vertical and longitudinal multi-layer graphene has been successfully grown on the substrate by a catalytic CVD using a new annealing method called "Heat-Beam (HB) CVD". We demonstrate its high potential of this method as a unit of a large–scale mass production machine.

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