Extended Abstracts of the 2012 International Conference on Solid State Devices and Materials, Kyoto, 2012, pp682-683

# Dependence of field effect mobility of CVD graphene on its grain size

Katsunori Yagi[1], Ayaka Yamada[1], Kenjiro Hayashi[1], Naoki Harada[1], Masahiro Tsukahara[2], Shinichi Ogawa[3], Shintaro Sato[1] and Naoki Yokoyama[1]

[1]Collaborative Research Team Green Nanoelectronics Center (GNC), AIST
[2]Innovation Center for Advanced Nanodevices (ICAN), AIST
[3]Nanoelectronics Research Institute (NRI), AIST
West 7A, 16-1 Onogawa, Tsukuba, Ibaraki 305-8569, Japan
Phone: +81-29-849-1555 Fax: +81-29-849-1186 E-mail: katsunori-yagi@aist.go.jp

## 1. Introduction

Graphene, two-dimensional honeycomb carbon lattice, is known to exhibit extremely high carrier mobility. However, grain boundaries in graphene adversely affect its electrical properties [1, 2]. Moreover, graphene formed on metal surfaces by chemical vapor deposition (CVD) should be transferred to an insulating substrate for device fabrication, and such a transfer process often contaminates and/or damages the graphene, deteriorating its electrical properties. In this study, we investigate the dependence of field-effect mobility in CVD-graphene on its grain size. We also investigate possible reasons for variation in the field effect mobility often observed in the experiments.

## 2. Experiment

Graphene was formed on a Cu film deposited on a 200-mm $SiO_2$/Si wafer using $C_2H_4$ or $CH_4$ diluted by Ar/$H_2$ as the source gas. The total gas pressure was 1 kPa, and the growth temperature was 860ºC, which was measured with a pyrometer. The thickness of Cu films was 500 nm or 1000 nm. The substrate was first annealed for 20 min in Ar/$H_2$ mixture (10:1), and $C_2H_4$ or $CH_4$ was then added for growth. The growth time and the partial pressure of hydrocarbons were changed, as shown in Table I. The graphene was analyzed by selected-area electron diffraction in transmission electron microscopy (TEM-SAED) and electron diffraction in scanning transmission microscopy (STEM-ED).

Table I Growth conditions of graphene

| Condition | Source | Partial pressure(Pa) | Growth period(min) |
|---|---|---|---|
| A | $C_2H_4$ | 3.9 | 0.5 |
| B | $C_2H_4$ | 0.59 | 4 |
| C | $C_2H_4$ | 0.079 | 60 |
| D | $CH_4$ | 0.68 | 60 |

The synthesized graphene was transferred to a $SiO_2$/Si wafer for device fabrication using the conventional transfer process. Briefly, a graphene/Cu/$SiO_2$/Si wafer was first spin-coated with photoresist. The wafer was then dipped in buffered HF to remove the $SiO_2$ layer, resulting in separation of the photoresist/ graphene/Cu film from the Si wafer. The Cu film was then removed in a $FeCl_3$ solution, and the graphene with the photoresist was placed on a $SiO_2$/Si wafer. The photoresist was then removed by immersing the wafer in acetone, resulting in graphene placed on the wafer.

Back-gate transistors were then fabricated using CVD-graphene on a $SiO_2$ (90nm)/Si wafer. In order to pattern graphene channels, the conventional photolithography process and $O_2$-plasma etching were used. The channel width and length ranged 0.5-20μm and 1-12μm, respectively. Source and drain electrodes were formed by electron-beam evaporation of Pd at both ends of the patterned graphene. The thickness of the electrodes was ~20 nm. The fabricated devices were observed by scanning electron microscopy (SEM; Hitachi SU8040)

Before measurements of electrical properties, the samples were annealed at 450ºC for 1 hour in vacuum to remove possible residues. The electrical properties were then measured in vacuum with semiconductor device analyzers (Agilent 4155C, B1500A).

## 3. Results and Discussion

The grain sizes of graphene were analyzed by TEM-SAED. The typical SAED patterns with an aperture size of 3 μm for the conditions A-D are shown in Fig.1.

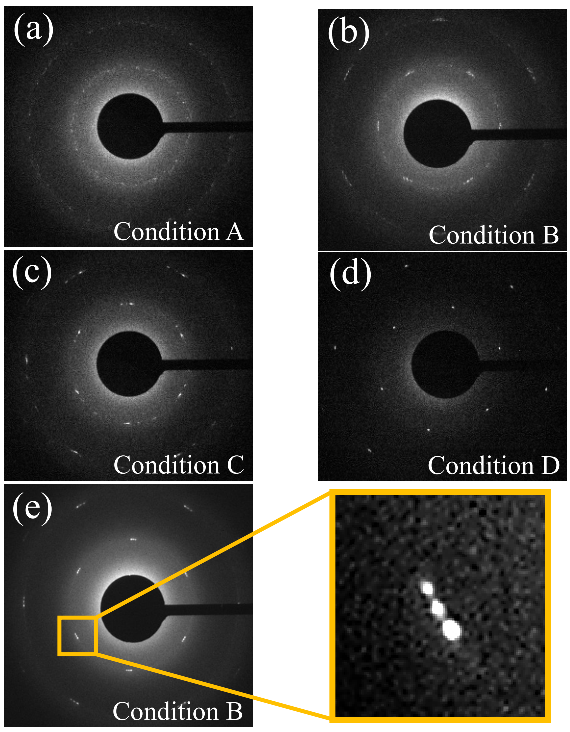


Fig. 1 TEM-SAED patterns of graphene samples grown under conditions A-D.

The approximate grain sizes estimated from TEM-SAED and STEM-ED (not shown) [3] are shown in Table II. In general, the lower the partial pressure is, the larger the grain size becomes. However, the grain size seems to saturate below a certain partial pressure. It was also found that, at the same partial pressure, a larger grain size was obtained with $CH_4$ than with $C_2H_4$.

Table II Grain size of graphene for each growth condition

| Condition | A | B | C | D |
|---|---|---|---|---|
| Grain size (nm) | 200-300 | 1000-1500 | 1000-1500 | 4000-5000 |

Field effect mobility was estimated from the transconductance of back-gate transistors. Mobility values as well as sheet resistances for each growth condition are shown in Fig. 2. They were obtained from two-terminal measurements, and therefore include the effects of the contact resistances between graphene and the source/drain electrodes. As can be seen in Fig. 2, it can be said that, overall, the larger the grain size is, the larger the mobility becomes. This is explained by the fact that channels made of graphene with larger grains are less likely to have grain boundaries that deteriorate their electrical properties [1,2]. Incidentally, we also performed four terminal measurements for graphene samples for condition D to exclude the contact resistances [1], and obtained the field-effect mobility as high as ~3,000 $cm^2/Vs$.

Of particular interest is that the mobility for condition B is higher than that for condition C, while the grain sizes are similar to each other. We made devices using graphene samples for conditions B and C a few times and obtained the same trend. In fact, graphene obtained under condition B sometimes gave a diffraction pattern shown in Fig. 1(e), which indicates that graphene are polycrystalline, but consists of aligned grains. Therefore, the difference between conditions B and C might be related to the alignment of grains. This issue, however, should be further investigated.

Mobility data shown in Fig. 2 are scattered. The scattered data are not actually explained by their possible dependence on the channel length and width. One possible reason for this scattering is fractures in graphene channels. Shown in Figs. 3(a) and (b) are SEM images of graphene channels with high mobility and low mobility, respectively. The graphene channel shown in Fig. 3(b) have a fracture in the channel from the lower left to the upper right, while that in Fig. 3(a) looks fine. Not only fractures but also wrinkles in the graphene channel seem to affect the mobility. Shown in Fig. 3(c) is a graphene channel with a lot of wrinkles. The mobility of the channel in Fig. 3(c) is actually one third of that in Fig. 3(a). We are planning to measure the temperature dependence of conductance in graphene channels with wrinkles to better understand their effects.

## 4. Conclusions

The grain sizes of CVD graphene samples were estimated and the dependence of field-effect mobility on the grain size was investigated. It was found that, overall, the larger the grain size is, the higher the mobility becomes. However, we also obtained the results suggesting that the grain alignment affected the mobility. It was also found that not only fractures but also wrinkles in graphene channels adversely affected the electrical properties.

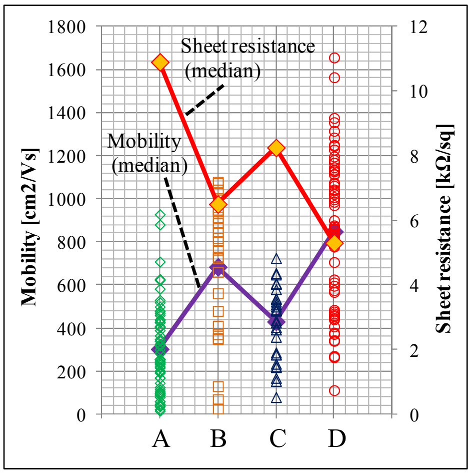


Fig. 2. Mobility of graphene channels grown under conditions A-D (hollow symbols; including the contact resistance). Medians of mobility and sheet resistance values are also shown (symbols with lines).

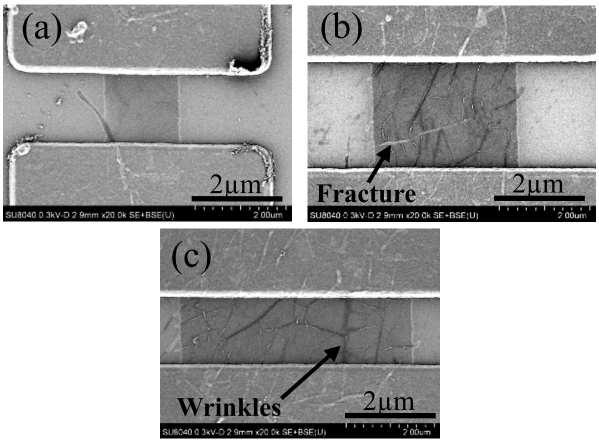


Fig. 3. SEM images of graphene (condition A) channels with mobility of (a) 900 cm2/Vs, (b) 16 cm2/Vs, and (c) 300 cm2/Vs.

## Acknowledgements

This work was supported by JSP through the "FIRST Program," initiated by CSTP, Japan. This work was partly conducted at the Nano-Processing Facility supported by ICAN, AIST, Japan.

## References

[1] H. S. Song, S. L. Li, H. Miyazaki, S. Sato, K. Hayashi, A. Yamada, N. Yokoyama and K. Tsukagoshi: Scientific Reports **2** (2012) 337
[2] Q. Yu, *et al.*: Nature Mater **10** (2011) 443-449
[3] K. Kim *et al.*: ACS Nano **5** (2011) 2142