Extended Abstracts of the 2012 International Conference on Solid State Devices and Materials, Kyoto, 2012, pp1168-1169

Annealing Condition Optimization of Sputtered Amorphous Carbon for Large-grain, Multi-layer Graphene

Motonobu Sato[1], Haruhisa Nakano[1],Makoto Takahashi[1], Takayuki Muro[2], Yuji Takakuwa[3], Shintaro Sato[1], Mizuhisa Nihei[1] and Naoki Yokoyama[1]

[1]Collaborative Research Team Green Nanoelectronics Center (GNC)/
National Institute of Advanced Industrial Science and Technology (AIST), 16-1 Onogawa, Tsukuba 305-8569, Japan
[2]Japan Synchrotron Radiation Research Institute (JASRI), 1-1-1 Kouto, Sayo-cho, Sayo-gun 679-5198, Japan
[3]Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan
Phone: +81-29-849-1578, Email: motonobu.sato@aist.go.jp

Abstract

In order to obtain large-grain, multi-layer graphene (MLG), we have investigated the effect of a catalyst and annealing ambient gas on the growth of MLG fabricated by post-annealing sputtered amorphous carbon. Higher quality graphene sheets stacked parallel to the SiO_2/Si substrate plane were obtained when using a Co catalyst layer and annealing in N_2 ambient gas. Moreover, the grain size of the carbon film was measured to be about 400 nm. By comparing these results with the results obtained with highly oriented pyrolytic graphite (HOPG), we speculated that the grain boundary is one of the reasons why the resistance of our film is still high.

1. Introduction

Carbon-based materials, such as carbon nanotubes (CNTs) and graphene nanoribbons (GNRs), have been studied as interconnect materials because they have lower resistivity [1], higher thermal conductivity [2], and intrinsically higher current-carrying capacity [3] than Cu.

In the past, we reported that the resistivity of sputtered amorphous carbon on dielectric layers without catalysts decreases as the annealing temperature increases, and that this decrease in resistivity can be attributed to an increase in sp^2 bonding corresponding to the formation of a graphene sheet [4]. Recently, we have succeeded in fabricating multilayer graphene (MLG) by annealing a sputtered carbon film with a catalyst layer [5]. And it is found that the MLG wire can sustain a high current density of 10^7 A/cm^2 which is over one order of magnitude higher than that of a Cu wire [6].

In this study, we investigated the effect of a catalyst and annealing ambient gas on the growth of MLG fabricated by post-annealing sputtered amorphous carbon.

2. Experimental

We used the magnetron sputtering method to deposit carbon and catalyst films onto a SiO_2/Si substrate at room temperature. The structures of the deposited films were Catalyst/C(30nm)/SiO_2/Si.

To investigate the effect of a catalyst, carbon films with various catalyst layers (Co, Pt, Ni, Au, or Cu) were annealed in N_2 flow gas at annealing temperatures of 800°C for 30min. And to investigate the effect of annealing ambient gas, carbon films with a Co catalyst layer were annealed in various flow gases (N_2, vacuum, Ar, or H_2) at annealing temperatures of 800°C for 30min.

The nanostructures of the films were analyzed by bulk-sensitive hard X-ray photoelectron spectroscopy (HAXPES) using BL47XU at SPring-8, X-ray diffraction (XRD) using Cu Kα radiation and electron backscatter diffraction pattern (EBSP).

3. Results and Discussion

Figure 1 shows the components of HAXPES C1s spectra in a carbon film with various catalyst layers after annealing at 800°C in N_2 flow gas. In the case of annealing with Co, Pt, or Ni, the ratio of sp^2 is almost the same and higher than that of Au or Cu. Conversely, in the case of annealing with Au or Cu, the ratio of amorphous, defects, and C-H components is higher than that of Co, Pt, or Ni.

Figure 2 shows XRD patterns (θ-2θ) of a carbon film with various catalyst layers after annealing at 800°C in N_2

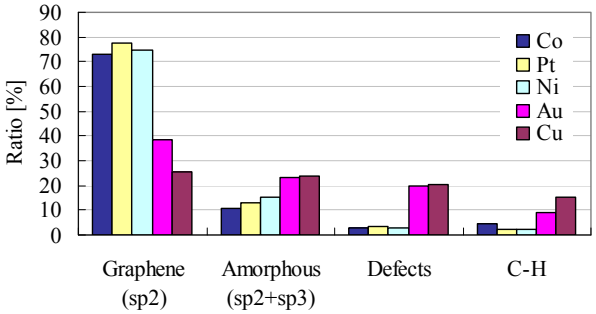

Fig. 1 Components of HAXPES C1s spectra in carbon film with various catalyst layers after annealing at 800°C in N_2 flow gas

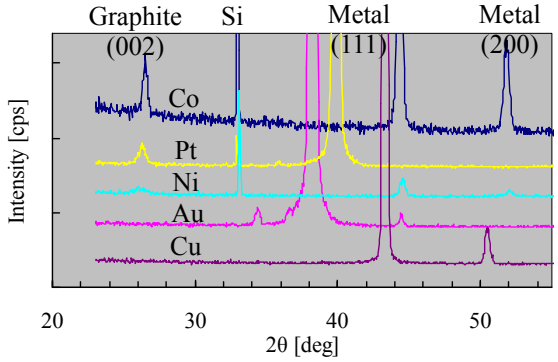

Fig. 2 XRD patterns (θ-2θ) of carbon film with various catalyst layers after annealing at 800°C in N_2 flow gas

flow gas. The carbon film with a Co, Pt, or Ni layer has a (002) peak of graphite. In particular, the intensity of that with Co is stronger than that of others. Therefore, these results suggest there are more graphene sheets stacked parallel to the SiO_2/Si substrate plane when using a Co catalyst than when using a Pt or Ni one, even though the sp^2 ratio is the same.

Next, we investigated the effect of annealing ambient gas using a Co catalyst. Figure 3 shows the components of HAXPES C1s spectra in a Co/C film after annealing at 800°C in various ambient gases. In the case of annealing in N_2, or a vacuum, the ratio of sp^2 is almost the same and higher than that of Ar or H_2. Conversely, when annealing in Ar or H_2, the ratio of amorphous, defects, and C-H components is higher than that of N_2 or a vacuum.

Figure 4 shows XRD patterns (θ-2θ) of a Co/C film after annealing at 800°C in various ambient gases. The annealed film in N_2 or a vacuum has a (002) peak of graphite. In particular, the intensity of that in N_2 is stronger than that in other gases. Therefore, these results suggest that more graphene sheets are stacked parallel to the SiO_2/Si substrate plane when using N_2 compared with annealing in a vacuum, even though the sp^2 ratio is the same.

Figure 5 shows the resistivity of a carbon film after removing Co of an annealed Co/C film. The resistivity of carbon when annealing in N_2 is the lowest, and depends on the (002) intensity of graphite in the XRD result. However, the resistivity of the film is one order of magnitude higher than that of HOPG.

Figure 6 shows the EBSP result of HOPG and Co/C film after Co removal. It is found that the grain size of the carbon film is about 400 nm, which is one order of magnitude smaller than that of HOPG. The resistance across the grain boundary is one order of magnitude higher than that within the grain [7]. Then, it is considered that the grain boundary is one of the reasons why the resistance of our film is higher than that of HOPG. To further improve the annealing conditions, it is necessary to enlarge the grain size so as to decrease the resistivity of MLG.

4. Conclusion

We optimized the catalyst and annealing ambient gas for obtaining large-grain, multi-layer graphene by post-annealing sputtered amorphous carbon. Measurements of the grain size also suggest that the grain boundary is one of the reasons why the resistance of our film is still high.

Acknowledgements

This research is supported by the Japan Society for the Promotion of Science (JSPS) through its Funding Program for World-Leading Innovative R&D on Science and Technology (FIRST Program).

References

[1] A. Naeemi and J. D. Meindl: Proc. of the 11th IEEE IITC (2008) 183
[2] N.Srivastava, R. V. Joshi, and K. Banerjee: IEDM (2005) 249
[3] B. Q. Wei, R. Vajtai and P. M.Ajayan: Appl. Phys. Lett. 79 (2001) 1172
[4] M. Sato, S. Ogawa, M. Inukai, E. Ikenaga, T. Muro, Y. Takakuwa, M. Nihei and N. Yokoyama: IITC/MAM (2011) 145
[5] M. Sato, M. Inukai, E. Ikenaga, T. Muro, S. Ogawa, Y. Takakuwa, H. Nakano, A. Kawabata, M. Nihei and N. Yokoyama: JJAP 51 (2012) 04DB01
[6] M. Sato, M. Takahashi, H. Nakano, T. Murakami, A. Kawabata, M. Nihei and N. Yokoyama: IITC (2012)
[7] O. Yu, et al., : Nature Mater. 10 (2011) 443

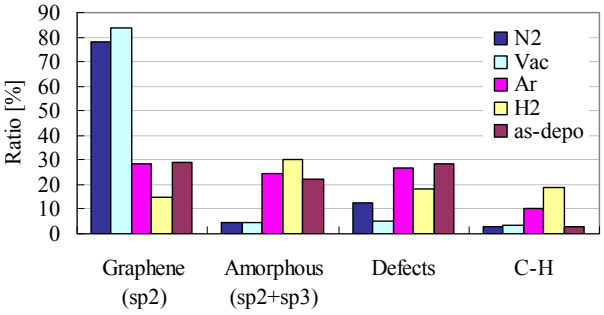

Fig. 3 Components of HAXPES C1s spectra in Co/C film after annealing at 800°C in various ambient gases

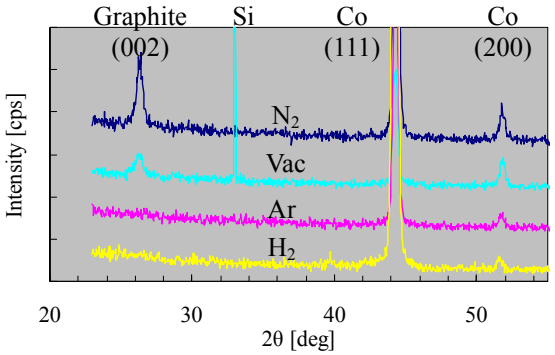

Fig. 4 XRD patterns (θ-2θ) of Co/C film after annealing at 800°C in various ambient gases

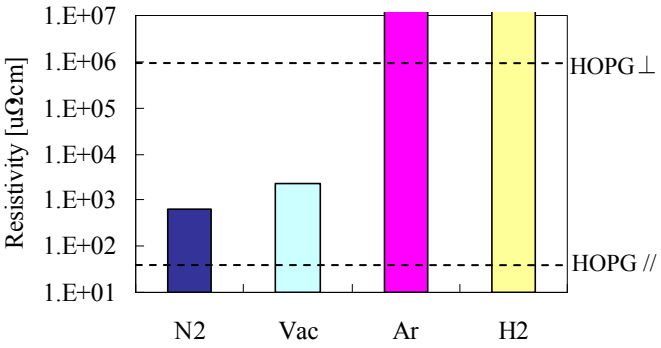

Fig. 5 Resistivity of a carbon film after removing Co of a Co/C film annealing at 800°C in various ambient gases

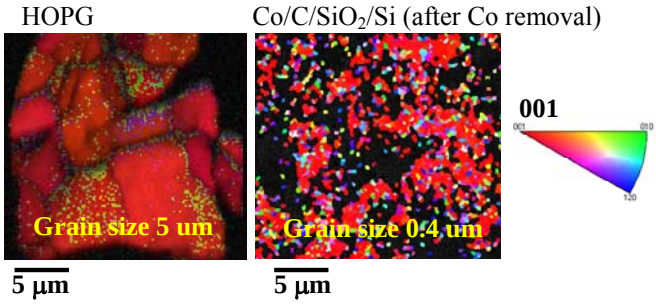

Fig. 6 EBSP of HOPG and Co/C film after Co removal