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Graphene as a Template Layer for the Growth of Ga-Based Compound Materials

Freddawati Rashiddy Wong[1], Nur Suhaili Abdul Aziz[1], Kanji Yasui[2] and Abdul Manaf Hashim[1,*]

[1] Malaysia-Japan International Institute of Technology, Universiti Teknologi Malaysia,
Jalan Semarak, 54100 Kuala Lumpur, Malaysia
*E-mail: abd_manaf@ic.utm.my
[2] Department of Electrical Engineering, Nagaoka University of Technology,
Kamitomioka-machi, Nagaoka, Niigata 940-2137, Japan

Abstract

We report the synthesis of gallium (Ga)-based compound materials on multi-layer graphene (MLG) on insulator by electrochemical deposition at room temperature (RT). The grown structures show grain-like thin film, and their thicknesses and sizes increase with the current density. Such grain-like morphology may due to the nature of graphene. The grown structures was found to be dominated by a mixture of cubic gallium nitride (c-GaN) and beta gallium oxide (β-Ga_2O_3) at current density above 2.5 mA/cm^2. The results open the breakthrough towards the selective growth of either c-GaN or β-Ga_2O_3 with further optimization of growth parameters.

1. Introduction

Up to this date, gallium-based compound materials such as gallium nitride (GaN) and gallium oxide (Ga_2O_3) have been used for wide range of applications in electronics and optoelectronics [1-2]. Graphene, a two-dimensional (2D) material, has also been proven as promising candidate for many applications such as device channel [3], transparent electrode [4], sensing membrane [5], etc, due to its extraordinary electrical, thermal, and mechanical properties, including a carrier mobility exceeding 10^4 cm^2/Vs and a thermal conductivity of 10^3 W/mK. Therefore, the integration of these materials on silicon (Si) should lead to the realization of the so-called heterogeneous integration on Si platform where it enables the excellent properties of these materials to be exploited in diverse sophisticated device applications. For practical application, these materials need to be isolated from Si by insulators such as silicon dioxide (SiO_2) or silicon nitride (Si_3N_4). It is very challenging to synthesize such materials including graphene with high quality directly on insulator. Recently, we show that graphene is a promising template or buffer layer to be used to grow silicon carbide (SiC) [6] and zinc oxide (ZnO) [7-8].

In this paper, we report the growth of Ga-based compound materials on insulator on Si by utilizing graphene as a template layer. It is noted that the growth of graphene on insulator is not considered in this study and commercially available transferred CVD grown graphene on SiO_2/Si substrate is used.

2. Experimental

The growth was carried out on MLG/SiO_2/Si substrate using a mixture of ammonium nitrate (NH_4NO_3) and gallium nitrate ($Ga(NO_3)_3$) dissolved in deionized (DI) water by a simple two terminal electrochemical deposition (ECD) at RT. The concentrations of NH_4NO_3 and $Ga(NO_3)_3$ solutions were fixed at 2.5 mol/L and 0.8 mol/L, respectively. These aqueous solutions were mixed in a teflon cell. A platinum (Pt) wire was used as an anode and a MLG/SiO_2/Si substrate as a cathode. The deposition was performed at different current densities ranging from 0.5 to 3.5 mA/cm^2 for 6 hrs.

3. Results and Discussion

Fig. 1(a)-(d) show the FESEM images of the grown structures at current densities of 0.5, 1.5, 2.5 and 3.5 mA/cm^2, respectively. The structures show grain-like morphologies with high degree of uniformity at current density above 2.5 mA/cm^2. Such morphology is believed to be originated from the polycrystalline graphene which is also having grain-like structures. The similar structure was also observed for the growth of SiC on graphene [6]. The cracking morphology is speculated to be caused by the uneven thicknesses of graphene layers. It clearly shows that the thicknesses and sizes of grain structures increase with the current density.

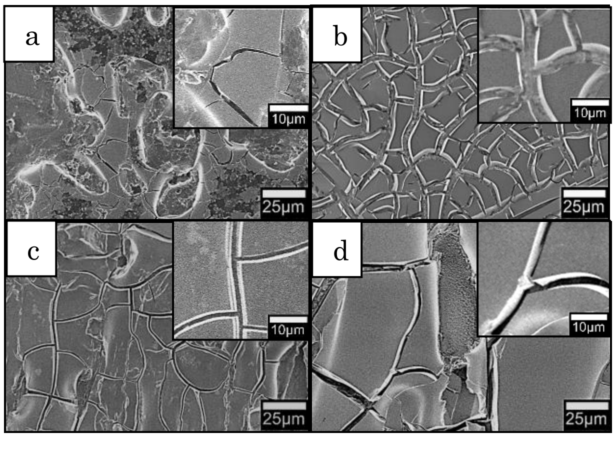

Fig. 1. FESEM images of grown structures at current densities of (a) 0.5, (b) 1.5, (c) 2.5 and (d) 3.5 mA/cm^2.

Fig. 2 shows the XRD spectra of the as-deposited structures and bare MLG/SiO_2/Si substrate. The 2θ peaks at 18.24˚ and 38˚ can be indexed to β-Ga_2O_3 in (20-1) and (31-1) planes, respectively (ICDD: 01-074-1776). While, the peaks at 40.41˚ and 58.81˚ can be indexed to c-GaN in (002) and (022) planes, respectively (ICDD: 01-088-2364). The intensities of the peaks for both Ga_2O_3 and GaN increase with current densities, which simply indicates the increase in thickness of the grown structures. The FWHM value of highly intense peaks for both Ga_2O_3 (20-1) and GaN (002) planes was estimated to be 0.80˚. In conclusion, it can be said that the grown structures were dominated by both Ga_2O_3 and GaN structures.

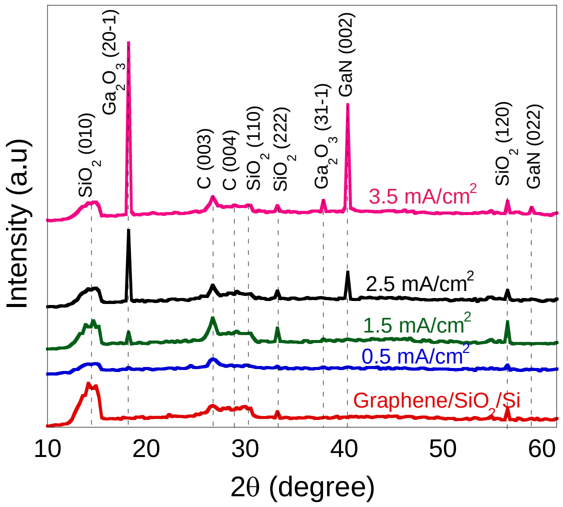

Fig. 2. XRD spectra of the grown structures and bare MLG/SiO_2/Si substrate.

Fig. 3 shows the measured FTIR pattern of the sample deposited at 3.5 mA/cm^2. Two bands with valley peaks at 464 and 665 cm^{-1} were observed which can be assigned to Ga_2O_3 (Ga-O bonding). While, the band with valley peak at 1082 cm^{-1} can be assigned to Ga-N bonding [9]. These results further confirmed the mixture of GaN and β- Ga_2O_3 structures.

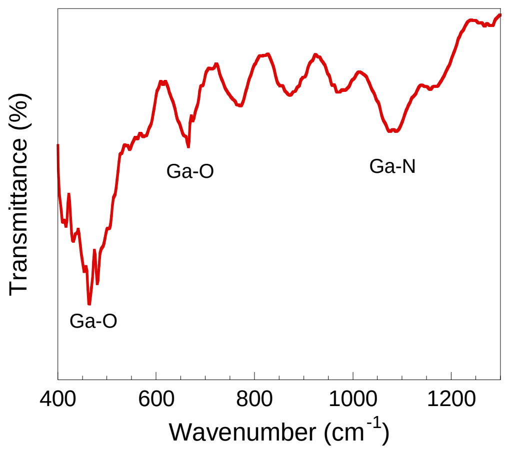

Fig. 3. FTIR spectrum of the grown structure at current density of 3.5 mA/cm^2.

The possible chemical reactions are summarized as follows:

$$NH_4NO_3 \rightarrow NH^{4+} + NO^{3-} \quad (1)$$
$$NH^{4+} + NO^{3-} \rightarrow NH_3 + HNO_3 \quad (2)$$
$$Ga(NO_3)_3 + H_2O \rightarrow Ga^{3+} + 3NO^{3-} \quad (3)$$
$$Ga(NO_3)_3 + NH_3 \rightarrow GaN + 3HNO_3 \quad (4)$$
$$Ga^{3+} + 2H_2O \rightarrow GaOOH + 3H^+ \quad (5)$$
$$2GaOOH \rightarrow Ga_2O_3 + H_2O \quad (6)$$

Here, firstly, the positive ions of Ga^{3+} and NH^{4+} are being concentrated on the surface of a cathode, where their reactions will form the clusters of c-GaN. Then, the excessive positive ions of Ga^{3+} will react with the molecules of water and form the clusters of Ga_2O_3. These reactions continuously take place and lead to the growth of thin film. From these reactions, we believe that the structure either Ga_2O_3 or GaN can be selectively grown. For example, to grow GaN dominated thin film, the molarity of NH_4NO_3 need to be increased, so that the molecules of NH_3 can be increased to aggressively react with the excessive positive ion of Ga^{3+}.

3. Conclusion

The growth of β-Ga_2O_3 and c-GaN on insulator by utilizing graphene as template layer was achieved. The results show that the continuous thin film can be achieved if large area single crystalline graphene is used. The selective growth is believed to be achievable by manipulating the molarity of the electrolyte towards either Ga_2O_3 dominated or GaN dominated thin film structures.

Acknowledgements

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