Synthesis of SWCNTs and Graphene 3D hybrid structures by ACCVD

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Graphene and single-walled carbon nanotubes (SWCNTs), sp² hybridized honeycomb structures of carbon, are realized as the promising materials for the wide ranges of applications due to their high carrier mobilities, high electrical and thermal conductivities, and large surface areas [1, 2]. A high-quality 3D hybrid structure of graphene and SWCNT is highly desirable to further enhance their properties so as to use in various applications [3,4]. Here we attempted to address this issue by growing small-diameter SWCNTs onto CVD synthesized graphene.

Graphene sheets were synthesized on Cu foils by chemical vapor deposition (CVD) technique and transferred onto SiO₂/Si as reported elsewhere [5]. Then, Ir nanoparticles were deposited onto them by dip coating by using Ir-acetate solution [6], which were utilized as growth substrates. SWCNT growth was carried out at an optimized condition using ethanol gas as carbon source in ultra-high vacuum CVD (UHV-CVD) system. Optical microscope (OM), Raman spectrometer, FESEM, and XPS were employed for the characterization.

Figure 1(a) shows Raman spectra observed onto dark and bright contrast areas (named as 1 and 5 respectively) of optical micrograph (OM, Figure 1(c)) by employing four different lasers as indicated in respective color codes (see inset), and radial breathing



Figure 1. (a, b) Raman spectra of SWCNTs grown onto graphene sheets and SiO₂/Si (regions 1 and 5 are shown in OM image of Figure 1(c)) measured by four different lasers as indicated in inset. Figure 1 (d) shows FESEM image of grown SWCNTs.

mode (RBM) area is further magnified in **Figure 1(b)**. Presence of RBM and G⁻peaks in Raman spectra for regions 1 and 5 along with degenerated 2D band for region-5 indicates the successful growth of SWCNTs onto Graphene sheets. Based on the RBM peaks observed in typical Raman spectra taken from both graphene and SiO₂/Si area (**Figure 1(b)**), grown SWCNTs were confirmed to have diameters between 0.8 and 1.1 nm in reference to Kataura plot. FESEM images (**Figure 1(d)**) further confirms that grown SWCNTs were vertically aligned.

This work was partly supported by the Meijo University Nanomaterial Research Center. Part of this work was supported by JSPS KAKENHI Grant Number JP19H02563 and "Advance Research Infrastructure for Materials and Nanotechnology in Japan (ARIM)".

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