Enabling the low temperature CVD growth of graphene using Alloy Catalyst and graphene induced abnormal grain growth of Cu-Ag alloy

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Abstract

Graphene, a two-dimensional crystalline structured material has been spotlighted from many researchers because of its fascinating electrical, mechanical, optical, thermal properties.^[1-6] One of the major challenges for the practical application of graphene has been the synthesis of large scale and uniform films with higher quality at lower temperatures. New catalytic design for graphene synthesis is essential to lower the synthesis temperature and improve the uniformity of graphene. Here, we demonstrate that the use of Ag-plated Cu substrates is very useful to synthesize the high quality graphene films via chemical vapor deposition (CVD) of methane gas at the temperature as low as 900 °C. In addition, we investigated that the abnormal grain growth of Cu with more than 1 mm² of grain size was induced by the graphene synthesis and this phenomena was occurred only for the Cu-Ag alloy foil among various type of Cu foils.

Graphene was synthesized on the Cu foil and Ag plated Cu foil at 800°C, 900 °C and 1000 °C using thermal chemical vapor deposition system. The synthesized graphene was inspected by optical microscope and Raman spectroscopy for the comparison of synthesis quality with the coverage of graphene film and the optimization of growth condition. Transmission electron microscope (TEM) was employed to determine crystal quality and uniformity of graphene and its domain size. For the analysis of Cu foil after graphene synthesis, scanning electron microscope (SEM) with Energy dispersive x-ray spectrometer, Dynamic-secondary ion mass spectrometry (D-SIMS) electron backscatter diffraction (EBSD) were used.

The uniform graphene film with full coverage was synthesized on the Ag plated Cu foil at 900 °C and

this synthesis temperature was much lower as compared with the common synthesis temperature of graphene on Cu.^[7] Synthesis state of highly uniform monolayer graphene was confirmed from the mapping analysis by Raman spectroscopy and TEM analysis. Graphene synthesis was enhanced with the increasing of Ag plating thickness for the thin Ag plating, while non-uniformity of graphene was increased for the thick Ag plating. High quality graphene without defects or bilayer sites was achieved with optimization of Ag plating thickness. The plated Ag diffuses into Cu and the formation of uniform Cu-Ag alloy was demonstrated by various analyses and this optimized Cu-Ag alloy controlled the formation of multilayer nucleation, leading to the lower synthesis temperature with enhanced monolayer coverage. In addition, the abnormal grain growth of Cu into cube texture with mm² scale of giant grain size was observed only on Cu-Ag alloy with graphene synthesis. The ratio of (100) was exceeded more than 90 % for Cu-Ag with graphene synthesis and this phenomena was not occurred if one of the condition of Ag or graphene was insufficient. In this talk, we report this unusual phenomena in detail and the role of Ag for low temperature CVD growth of high quality graphene.

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Figures



Figure 1 Raman spectra of graphene synthesized on (a) Cu and (b) Ag200Cu at 800 °C, 900 °C and 1,000 °C. Optical microscopic images of graphene synthesized on (c) Cu (d) Ag200Cu at 900 °C for 40 min. Uniform and complete graphene film was synthesized for Ag200Cu at 900 °C.^[7]



Figure 2 EBSD orientation maps of Cu after graphene synthesis on Cu (a) and Ag200Cu (b) at 800 °C, 900 °C, 1,000 °C; Scale bars, 200 μm.^[7]