

プラズマを利用した高品質グラフェン合成と透明導電フィルム応用

High Quality Graphene Synthesis by Plasma Technique and Applications for Transparent Conductive Films

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1. Introduction

We developed the large-area microwave plasma chemical vapor deposition (CVD) of graphene for transparent electrode applications. This technique has been successfully combined with the roll-to-roll process to synthesize graphene on Cu substrates. Nucleation density of plasma CVD of graphene crystal is too high, which suppresses two-dimensional growth of graphene and leads to graphene flakes of several nanometer stacks in multiple layers. This causes low electrical conductivity of the synthesized graphene. The low concentration of carbon source is effective to reduce the nucleation density and to enlarge the crystal size. In this study, we utilize small amount of carbon delivered from Cu foil and/or the ambient in the reaction chamber as extremely low-concentration of carbon source and perform the synthesis of graphene of higher crystalline quality.

2. Experimental and Results

The carbon atoms precipitate on the Cu surface by the heat treatment of the Cu foil for 15 minutes at about 800°C. The heat treatment was performed by direct Joule heating of Cu foil. Then the copper foil was exposed to the hydrogen plasma for about 30 seconds under 5Pa to synthesize graphene on the foil. We did not use any carbon-contained gas such as CH₄, which is most significant difference from the conventional plasma CVD method of graphene. Fig.1 (a) shows the Raman spectrum (XploRa, HORIBA, spot size 1μm, 638nm) of synthesized graphene film. On the other hand, fig.1 (b) indicates Raman spectrum of graphene film synthesized by the plasma CVD using CH₄ as the carbon source as a reference. In fig.1 (a) the D band is much smaller and the 2D band is sharper and stronger than in fig.1 (b). These results indicate that the crystalline quality of graphene was successfully improved by using carbon atoms contained in the Cu foil.

The synthesized graphene on Cu foil was transferred to the quartz or silicon substrate. After that, the van der Pauw devices for Hall Effect measurements were fabricated using conventional photolithography, metal deposition and lift-off processes. The fabricated devices show that the

mobility is estimated to be more than 1000 cm²/Vs, which was dramatically improved from the previous plasma CVD using CH₄ as source gas.

3. Summary

We have developed a new synthesis method of graphene by plasma treatment using extremely small concentration of carbon source to enhance two-dimensional crystal growth. Raman spectroscopy and Hall Effect measurements revealed dramatically improvement of the crystalline quality and the electrical conductivity.

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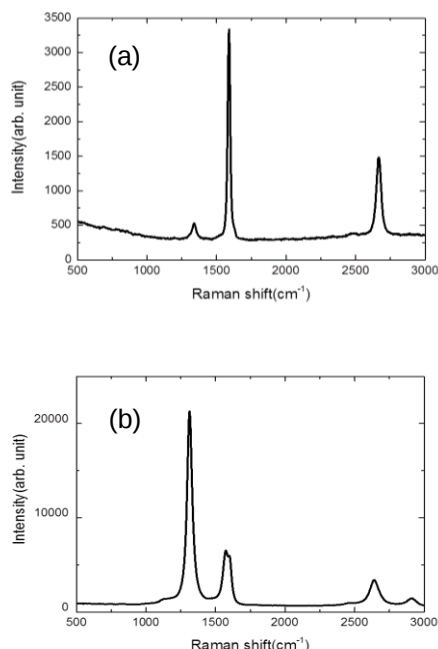


Fig.1 Raman spectra of graphene synthesized by plasma treatment of copper foil by using (a) extremely low concentration of carbon source, and (b) using CH₄ gas.