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Preparation of few-layered graphene using Graphite Intercalation Compounds (GICs)

1. Introduction

The preparation of the non-oxidezed graphene have been demanded for lomg time. Non-oxidized few-layered graphene is pepared by adding water to K-tetrahydrofuran-Graphite Intercalation Compounds (K-THF-GICs), which are consider to be ternary GICs. However, prepared graphene fails to achieve the large area required for pratical appricatios. The addition of an electron–withdrawing aldehyde may result in the fabrication of few-layered graphene on the carbon-based hexagonal netwoks by nucleophilic substitution reaction, forming thin and large-area graphene sheets. Furthermore, an aldehyde solvent containing long alkyl chains is preferable for the preparation of large-area few-layered graphene when compared with an aldehyde solvent containing short alkyl chains[1]. Therefore, we can considered that few-layered graphene can be prepared from alcohol or other solvents containing long alkyl chains. This study demonstrated the fabrication of non-oxidized few-layered graphene with a large area by the addition of a long alkyl chains aldehyde and alcohol soluition to the host K-THF-GICs.

2. Materials and methods

A K-THF solution was prepared by dissolving naphthalene in THF and adding potassium and by stirring for 30 min. Subsequently, K-THF-GICs were prepared by soaking the as-obtained graphite (grain size = 100 μm) in the prepared solution. Few-layered graphene was further prepared by the addition of decanal and decanol (having long alkyl chains) to the K-THF-GICs and stirring for 72 h. After stirring, supernatant and sediment were separated and the supernatant was centrifuged for obtaining few-layered graphene. The synthesized K-THF-GICs were characterized using XRD and the obtained graphene was characterized using TEM imaging and Raman spectroscopy respectively.

3. Result and discussion

Figs. 1 depicts the XRD patterns of the synthesized K-THF-GICs. The sequence peaks are contributed by stage-1 of the phase-A K-THF-GICs. In the phase A, the THF molecules that coordinate with potassium in the graphite layer are observed to be a perpendicular to the graphite layer. Few-layered graphene was prepared by the addition of decanal and 1-decanol to the K-THF-GICs. Figs. 2(a) and (b) represent the TEM micrographs of few-layered graphene obtained by the addition of decanal and 1-decanol to the K-THF-GICs, respectively. As revealed in both micrographs, few-layered graphene covered an area spanning dozens of square micrometers, which was larger than the area covered by that

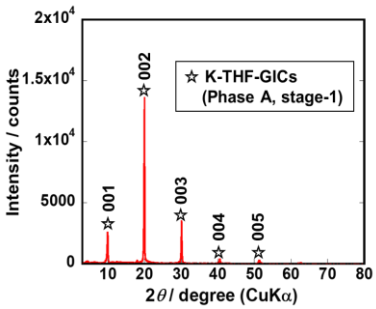

Fig.1 XRD patterns of K-THF-GICs

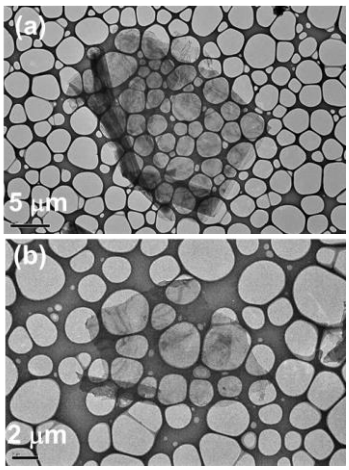

Fig.2 TEM micrographs of graphene obtained by the exfoliation of K-THF-GICs in (a) decanal solution and (b) 1-decanol solution.

prepared by the addition of the water to the K-THF-GICs (6.2 μm^2 on an average). Figs. 3 depicts the Raman spectra of as-obtained graphite and the products obtained from decanal and 1-decanol using proposed technique (i.e., by separating the precipitate in a supernatant). In the graphite spectrum (Figs. 3(a)), the two-dimensional (2D) band can be observed at approximately 2726 cm^{-1}, whereas the spectrum is the red-shifted to approximately 2693 cm^{-1} and 2701 cm^{-1} in the spectra of the decanal and 1-decanol products, respectively (Figs. 3(b) and (c), respectively). This red shift indicates the presence of five or fewer lamination layers in the graphene sheets. The insets in small panels (a), (b) and (c) of Figs. 3, denote the peak separation results of the 2D band in the graphite, decanal product, and 1-decanol product, respectively. The main peak was split into two peaks using approximate area ratios of the low-wavenumber peaks at 42, 57 and 80% in the 2D band after peak separation, respectively. There results indicate the presence of small number of layers in the decanal and 1-decanol products. Based on the aforementioned results, we can conclude that few-layered graphene can be prepared by adding decanal and 1-decanol (having long alkyl chains) to the K-THF-GICs.

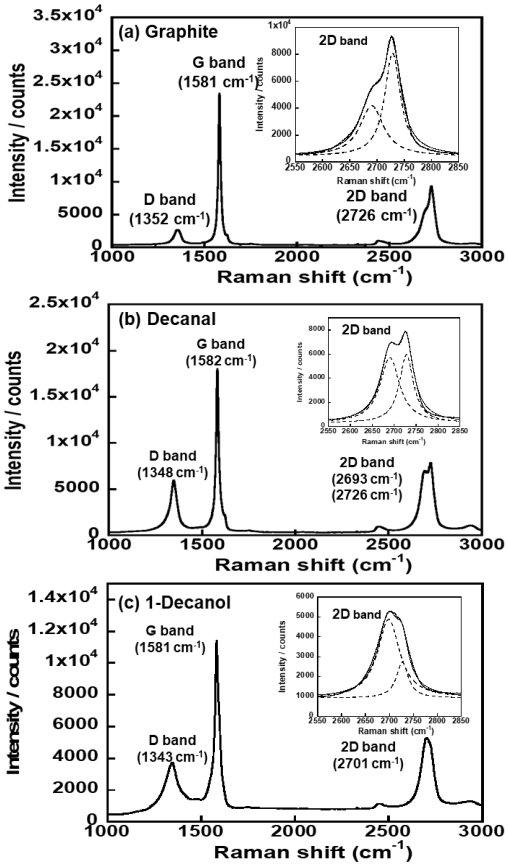

Fig.3 Raman spectra of (a) natural graphite and (b) precipitate in supernatant solution obtained K-THF-GICs exfoliated by addition of decanal solution, (c) 1-decanol solution.

Reference

[1] Yoshihisa Nanri, Hiroshi Yoshitani, Hiroji Fukui, Akira Nakasuga, Taro Kinumoto, Tomoki Tsumura, Masahiro Toyoda, 「Preparation of few-layered graphene using Graphite Intercalation Compounds (GICs)」, The 56th FNTG Society 3P-10, 2019