Supporting Information

Synthesis of graphene film from fullerene rods

Rajanish N. Tiwari,*^a M. Ishihara,^a Jitendra N. Tiwari^b and M. Yoshimura^a

Toyota technological Institute, 2-12-1 Hisakata, Tampaku-ku, Nagoya 468-8511, Japan

^bCenter for Superfunctional Materials, Department of Chemistry, Pohang University of Science

and Technology San 31, Hyojadong, Namgu, Pohang 790-784, South Korea.

*Corresponding author Rajanish N. Tiwari E-mail: <u>rajanisht@gmail.com</u> Fax: +81-52-809-1851 Tel: +81-52-809-1852

Experimental Section

The pressure was generated on prepared sandwich structure (Mo-C60-Ni-dielectric-Mo) by manually. The sandwich structure was fixed by screw. The dielectric material (SiO₂/Si) in sandwich structure should not be broken during fixing by screw. The melting point of Mo is very high (2623°C). The major role of Mo is to give support for fullerenes and Ni and to create pressure on the sandwich structure by screw. After generating pressure on structure, the synthesis process was carried out under low vacuum such as 1×10^{-1} Pa.

Results and discussion

In general, the solubility depends on three kinds of interaction such as solvent-solvent interaction, solvent-solute interaction and solute-solute interaction. Probably, such kind of interactions between fullerene and toluene molecules resulted unique morphology such as fullerene rod-like crystals in the toluene solvent [1]. The inset high-magnification SEM image in Figure 1S (A) shows that the size of fullerene rods-like crystal size are varied from ~2 to ~3µm. The cross-sectional SEM image in Figure 1S (B) shows the thickness of fullerene rod-like crystals. From the cross-sectional SEM image (low and high magnification), it is clear that fullerene rods-crystals ~ 2µm in thickness are deposited on the surface after drop method. Further, the fullerene-rod-like crystals were evaluated by Raman spectroscopy. Raman spectroscopy was performed using a REINSHAW system with a diode laser (wavelength = 532nm), which could be focused to a diameter of 1 µm for micromode operation. Intense Raman peaks in the range of 1000-2000 cm⁻¹ are shown in Figure 1S (C). All the multi Raman peaks at 1420, 1464 and 1587 cm⁻¹ are the typical characteristics of fullerene molecules. It seems that toluene is evaporated and only fullerene molecules are left on the surface for graphene synthesis. In addition, we used Rigaku XRD to evaluate the fullerene on Si surface at ambient temperature.

The XRD pattern in Figure 1S (D) shows that the diffraction peaks at $2\theta = 10.84$, 17.1 and 21.74 correspond to the interplanar spacings of fullerene {111}, {220} and {222}, respectively, indicating that polycrystalline fullerene rod–like crystals were deposited on the surface. It is also clear that the significant amount of fullerene molecules has only survived and toluene molecules either completely desorbed or presented in insignificant amount on the surface. Therefore, we did not notice any peak of toluene in Raman spectrum, as shown in Figure 1S (C).



Figure 1S (A) Plan-view SEM image; inset high magnification image (B) Cross-section SEM image; inset high-magnification image (C) Raman spectrum and (D) X-ray diffraction pattern of fullerene.



Figure 2S Plan-view SEM image of fullerene rods after 10 min annealing at 600 °C.



Figure 3S HRTEM image and their hexagonal FFT pattern show the formation of single layer graphene at 750 °C after 10 min annealing.



Figure 4S Transparency spectra of single layer graphene shows the average transmittance (450-800) is $\sim 91\%$.

Reference

.

[1] Y. Shen, J. Wang, U. Kuhlmann, P. Hildebrandt, K. Ariga, H. Mçhwald, D. G. Kurth and T.

Nakanishi, Chem. Eur. J. 2009, 15, 2763.