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Supplementary Material

Proton Conductivity of Graphene Oxide on Aging

Mohammad Razaul Karim,[A,B,D] *Md. Saidul Islam,*[A] *Nurun Nahar Rabin,*[A] *Ryo Ohtani,*[A] *Masaaki Nakamura,*[A] *Michio Koinuma,*[A] *and Shinya Hayami*[A,C,D]

[A] Department of chemistry, GSST, Kumamoto University, 2-39-1 Kurokami, Chuo-ku, Kumamoto 860-8555, Japan.
[B] Department of Chemistry, Shahjalal University of Science and Technology, Sylhet-3114, Bangladesh.
[C] Institute of Pulsed Power Science, Kumamoto University, -39-1 Kurokami, Chuo-ku, Kumamoto 860-8555, Japan.
[D] Corresponding authors. Email: krazaul@yahoo.com, hayami@sci.kumamoto-u.ac.jp

Experimental details

Synthesis of GO: GO was synthesized by Hummers' method, where 5.0 g powdered graphite, 5.0 g $NaNO_3$ and 250 ml 97% H_2SO_4 was cooled to 0 ºC in an ice bath. 15.0 g finely meshed $KMnO_4$ powder was slowly added with vigorous stirring. The temperature during stirring was maintained below 20 ºC. After 35 min, the mixture was warmed to 35 ºC for 40 min. and then 1 L. water was added slowly. The temperature increased gradually and was maintained around 95 ºC for another 30 min. Then 2 L water and 60 mL 30% H_2O_2 was added. The mixture was centrifuged at 4000 rpm for 10 min. The precipitate was washed one time with 5% HCl solution and 3 times with water. The precipitate was then dried at 70 ºC for 24 hours to obtain H-GO.

Preservation of GO. GO was preserved in a transparent glass bottle sealed at room temperature and was kept in a rack of the laboratory cabinet.

Characterization of GO: The structure and morphology of GO were characterized by using X-ray photoelectron spectroscopy (XPS), X ray diffraction spectroscopy (XRD), SEM images and RAMAN spectroscopy. Surface morphology of the samples were studied by a field-emission SEM (Hitachi High-Tech, SU-8000). An XPS instrument of Thermo Scientific, Sigma Probe was used for studying the X-ray photoelectron spectroscopy. A monochromatized X-ray source (Al Kα, hν = 1486.6 eV) and a discharge source (He I, hν = 21.2 eV) was used in the system. Pt substrate was used to determine the Fermi level in GO/Pt film. Vacuums better than 10^{-7} Pa was

ensured during the measurements. A hemispherical energy analyzer equipped with six channeltrons was used to detect the emitted electron. Study of Raman spectroscopy was performed using a micro Raman spectrometer (NRS-3100, Jasco, Japan) with a 532 nm excitation source at room temperature.

Measurement of Proton conductivity: Samples were compressed into pellets with an area of; both sides of which were attached to gold wire and gold paste. The sample Conductivity (σ) was calculated as, $\sigma = (1/R)\times(d/A)$ where, R, d and A are the resistance (radius of the semicircular curve of Nyquist plot), thickness and area of the samples. In Nyquist plots, the curves are fitted from the real (Z′) and imaginary parts (Z″) of the impedance as the frequency was varied. Temperature and relative humidity (RH) dependent impedances were recorded. Proton conductivities of the samples were measured by four-probe impedance/gain phase analyzing system using a Solaratron 1260/1296 in the frequency range from 1 to 10^6 Hz. 100 mg sample was mashed into powdered forms and mechanically compressed into coin like pellet of $1cm^2$ area and thicknesses of 0.1cm. Both sides of the pellet were attached to gold wire (50 μm diameter) with gold paste, obtained from Tanaka Kikinzoku Kogyo. Much care was taken to avoid connection of the tow faces. The Impedances with respect to the modulation of both the relative humidity (RH) and temperature using an incubator (SH-221, ESPEC) were recorded. For each measurement incubation time was 1 hour. The σ values (conductivity) were calculated using the equation, $\sigma = d/RA$, where d is the thickness of the pellet (the distance between the electrodes), R is the measured resistance and A is the area of the electrode. For all samples, ‘d’ and ‘A’ had the fixed value as 0.1 cm and 1 cm^2, respectively.