## **Supporting information**

## Supercritical Fluid Assisted Synthesis of N-doped Graphene Nanosheets and Its Capacitance Behavior in Ionic liquid and **Aqueous Electrolyte**

Marappan Sathish,\*<sup>*a,b*</sup> Satoshi Mitani,<sup>*a*</sup> Takaaki Tomai<sup>*a*</sup> and Itaru Honma\*<sup>*a*</sup>

<sup>a</sup>Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, 2-1-1, Katahira, Sendai 980 8577, Japan. <sup>b</sup>Functional Materials Division, CSIR-Central Electrochemical Research Institute, Karaikudi -630 006, India.

E.mail: marappan.sathish@gmail.com; i.honma@tagen.tohoku.ac.jp

GO nanosheets preparation: GO nanosheets were prepared by modified Hummers and Offeman's method reported elsewhere [1, 2]. Graphite powder (5 to 20 micrometer, obtained from Sigma-Aldrich) was used as starting materials. In a typical preparation, 1 g of graphite, 1 g of NaNO<sub>3</sub>, and 46 mL of H<sub>2</sub>SO<sub>4</sub> were stirred together in an ice water bath. Then, 6 g of KMnO<sub>4</sub> was slowly added. Once mixed, the solution was transferred to a  $35 \pm 5$  °C water bath and stirred for about 1 h, forming a thick paste. 80 mL of water was added to the above paste and the resulting solution was stirred for 30 min while the temperature was raised to 90  $\pm$  5 °C. Finally, 200 mL of water containing 6 mL of H<sub>2</sub>O<sub>2</sub> was added, the color of the solution turns from dark brown to yellow. The warm solution was then filtered and washed with 400 mL of water. The filter cake was then dispersed in water with mechanical agitation and mild sonication, giving a solution of exfoliated GO.

## N-doped graphene synthesis

In a typical synthesis, N-containing organic compounds, 0.5g of Ethylenediamine or Melamine or hexamethylenetetramine was dissolved in ethanol 10 mL solution. The above solution was added into GO solution (~35 mg/mL) and stirred for 30 min followed by ultrasonication for 30 min at room temperature. The ratio of GO and N-containing organic compound is maintained as 1:1 wt %. Then, the above mixture (5 mL) was loaded into stainless steel reactors of total 10 mL capacity and the reactors were placed into pre-heated (400 °C) furnace with constant shaking (within a specifically designed tubular furnace, AKICO, Japan). After 30 min, the reactors were quenched in an ice–water tank and the product was collected after several washings with distilled water and ethanol, and dried at 70 °C in a vacuum oven for 12 h.

*Reduced N-doped graphene nanosheets preparation.*0.5 g of N-doped graphene nanosheetes obtained in the above process was redispersed into 30 mL of distilled water and placed into a hot plate at 90°C and stirred. 0.1 ml of hydrazine hydrate was added and stirred for another 2h at 90°C. Then, the resulting sample was filtered, washed with distilled water and dried at 60°C for overnight.

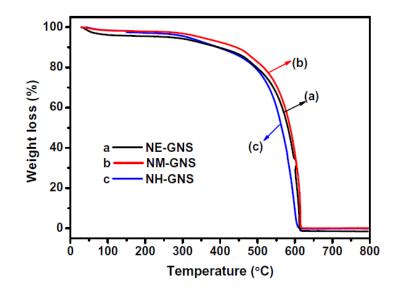


Fig S1. TG profile of (a) NE-GNS, (b) NM-GNs and (c) NH-GNS

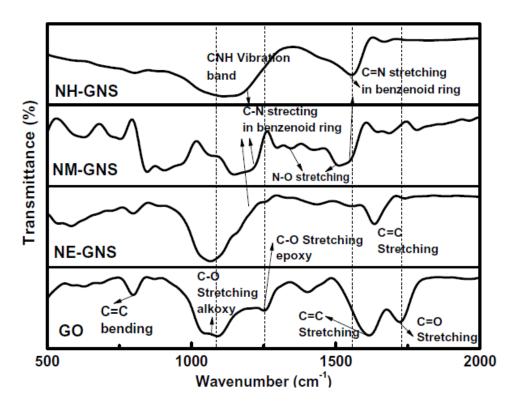


Fig S2. FT-IR Spectra of GO, NE-GNS, NM-GNS and NH-GNS

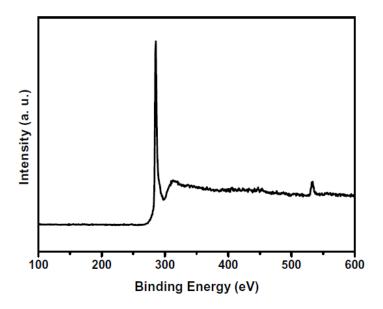


Fig S3. XPS spectrum of RGO nanosheetes prepared in supercritical fluid assisted method

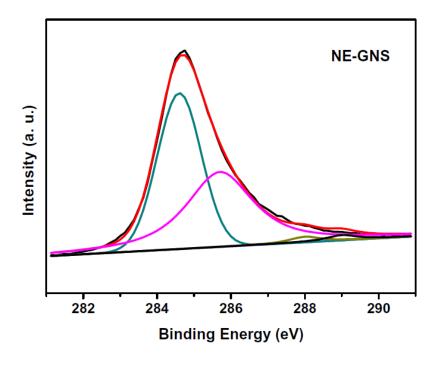


Fig S4. XPS spectra of NE-GNS

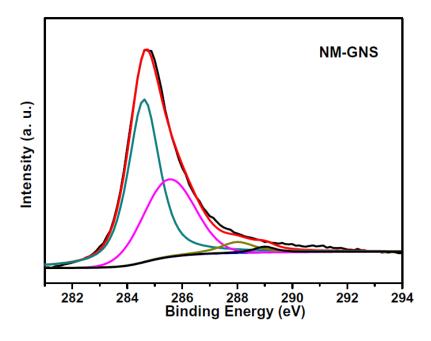


Fig S5. XPS spectra of NM-GNS

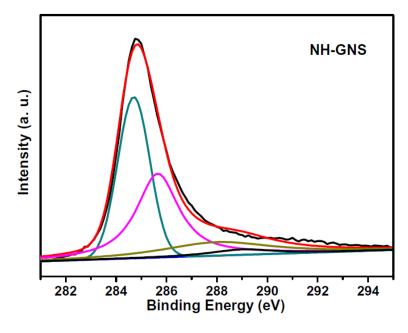


Fig S6. XPS spectra of NH-GNS

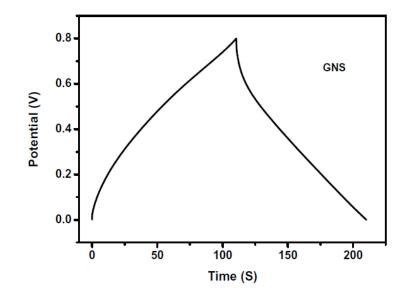


Fig S7. Galvanostatic charge-discharge profile of GNS prepared in supercritical fluid assisted method