Supporting Information

Synthesis and Electrochemistry of Highly Pseudocapacitive Carbon Nano Onions aka Multilayer Fullerenes and its MnO₂ nanocomposite

Vedi kuyil Azagan Ma, Mukta V. Vaishampayan, Manjusha V. Shelke\textsuperscript{a,b,c,*}

\textsuperscript{a}Physical and Materials Chemistry Division, CSIR-National Chemical Laboratory, Pune-411008, MH, India.
\textsuperscript{b}CSIR-Network Institute for Solar Energy, CSIR-National Chemical Laboratory, Pune-411008, MH, India.
\textsuperscript{c}Academy of Scientific and Innovative Research (AcSIR), Anusandhan Bhawan, 2 Rafi Marg, New Delhi-110 001

1. Comparative CV of CNOs, MnO₂/CNOs and carbon paper:

![Comparative cyclic voltamogram](image)

**Figure S1.** Comparative cyclic voltamogram shows the contribution of bare carbon paper which was used as the current collector at the scan rate of 5mV/s.
2. **Surface area analysis:**

![Graph showing nitrogen sorption isotherms and pore size distributions of CNOs.](image)

**Figure S2.** Nitrogen sorption isotherms at 77 K and the Inset shows pore size distributions of CNOs.

By using Brunauer-Emmett-Teller (BET) theory and Density Functional Theory (DFT) on the collected nitrogen-adsorption isotherms, pore size and the surface area distributions were calculated. For N$_2$-sorption measurements, Autosorb-iQ automatic volumetric instrument was performed at 77 with pressures in the range 0–760 Torr. The calculated average pore size of CNOs chain structure is ranging from 1.2-1.4 nm. CNOs exhibit VI isotherm and it has surface area of 486 m$^2$ g$^{-1}$. 
3. **HRTEM images of CNOs:**

![HRTEM images of CNOs](image1)

**Figure S3.** HRTEM images of CNOs showing (a) completed ring of multilayer fullerene and (b) fused fullerene rings

4. **TEM images of thermal carbon black:**

![TEM images of thermal carbon black](image2)

**Figure S4.** (a,b,c) Shows the HRTEM images of as-collected thermal carbon black (d) shows the SAED pattern of the CNO, the inter atomic distance calculated is 0.34nm; corresponding to (002) plane of carbon.
5. *Thermal gravimetric analysis (TGA)*:

![Thermogravimetric analysis (TGA) of CNOs and its composite MnO$_2$/CNOs before annealing at 800°C.](image)

**Figure S5.** Thermogravimetric analysis (TGA) of CNOs and its composite MnO$_2$/CNOs before annealing at 800°C.

For MnO$_2$/CNOs composite 5% weight loss has been observed between 30 and 140 °C that is attributed to the liberation of adsorbed water molecules from the composites. Low onset potential shows pyrolysis of organic residue. Further weight loss of 60% up to 445 °C corresponds to the loss of organic residues in CNOs. In addition, we have observed a final weight loss of 1% after 880 °C is due to the conversion of MnO$_2$ to Mn$_3$O$_4$.[Xiaofeng Xie, Lian Gao, ‘Characterization of a manganese dioxide/carbon nanotubes composite fabricated using an in situ coating method’, *Carbon*, 45, (2007) 2365–2373.]
6. **XPS of MnO2/CNOs composite:**

![XPS spectra of MnO2/CNOs composite](image)

**Figure S6.** XPS spectra of MnO2/CNOs composite.

The XPS measurements have been taken to study the components and oxidation states of CNOs and MnO2/CNOs composite. In figure (a) the binding energies 644.2 eV and 654.2 eV corresponding to Mn 2p band for the binding energies 2p 1/2 and 2p 3/2 which proves that the Mn in the composite is having oxidation state IV. The XPS of O 1s the has been deconvoluted into three components (fig b); the binding energy at 529.86 eV gives the evidence for Mn-O-Mn bonds for the tetravalent oxide which has highest intensity among others and the another binding energy at 531.19 eV shows that the hydroxide group has been bound to manganese Mn-OH; the binding energy at 532.48 eV corresponds to H-O-H bonds.

7. **SEM images of precursor ghee, thermal carbon black and CNOs:**

![SEM images](image)
**Figure S7.** SEM images of (a) ghee (butter oil), (b) as collected thermal black carbon and (c), (d) CNOs after annealing at 800°C.