Supporting Information

Carbon nanocoils for multi-functional energy applications

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S1: Preparation of catalyst for the synthesis of MWNC:

MmNi3 alloy target has been prepared by arc melting the constituent elements in a stoichiometric ratio under argon atmosphere. Alloy thin films have been deposited on carbon fabric by reactive RF magnetron sputtering technique by applying suitable RF power for 15 min. at 300 °C. The as-grown MmNi3 alloy thin film catalyst has been hydrogenated to a maximum storage capacity using a specifically built high pressure Sieverts apparatus. Fine particles of alloy thin film was obtained after several cycles of hydrogenation/dehydrogenation and the scanning electron microscopy (SEM) was used to investigate the morphology of alloy thin films and alloy hydride thin films.

S2: Synthesis of MWNC:

Alloy hydride thin film was directly placed in a quartz boat and kept at the center of a quartz tube, which was placed inside a tubular furnace. Hydrogen (50 sccm) was introduced into the quartz tube for 1 h at 500 °C, and then the furnace was heated up to 700 °C, followed by the introduction of acetylene for 30 min with a flow rate of 70 sccm. Acetylene flow was stopped and the furnace was cooled to room temperature. Argon flow was maintained throughout the experiment. In water assisted CVD technique, an acetylene gas has been passed through the water bubbler before reaching it to the reaction zone. SEM, TEM (JEM 200 FX II) measurements were carried out for characterizing these coiled nanotubes.
**Fig. S1:** Raman spectra of as-grown MWNCs prepared by pyrolysis of acetylene over alloy hydride thin film catalyst coated carbon fabric.

**Growth mechanism of MWNC:**

During our growth process, a carbon fabric which consists of a backing layer, coated with MmNi₃ alloy thin film has been used. Carbon black (Vulcan XC-72) was treated at 600°C for 3 h to remove organic matter. Next, the pretreated fabric was mixed with polytetrafluoroethylene (PTFE) and isopropyl alcohol in a supersonic mixer. Finally, this mixture was applied on wet proofed carbon fabric and dried at 300 °C. Then, MmNi3 thin film coated carbon fabric has been subjected to hydrogen absorption/desorption cycles, which has resulted in the exposure of pretreated carbon with PTFE to the hydrocarbon. As PTFE undergoes decomposition at temperatures of ~ 350°C, the pyrolysis of PTFE leads to liberation of different complex products. These liberated gases may cause defects to the growing nanotubes and there by leading to the formation of coil structure to the nanotube. As there is a probable chance for the evaporation of carbon black from the carbon fabric, the formation of MWNC would have also been assisted by the carbon vapour coming out from
the carbon fabric. Hence, the overall growth mechanism of MWNC is assisted by multiple carbon sources having different kinetic energies. The difference in kinetic energy leads to the formation of defects in the growing nanotubes which results in the formation of coiled structure.

**S3: Preparation of Pt/MWNC and Pt-Ru/MWNC:**

Purified MWNC were ultrasonicated in concentrated nitric acid for 3 h. After the sonication procedure, MWNCs sample were washed with de-ionized water several times and dried in air for 30 min at 100°C. The dried sample was ultrasonicated in 10 ml of acetone for 1 h and then 0.075 M H$_2$PtCl$_6$ solution was added slowly during stirring. After 12 h, the mixture was reduced by adding reducing solution containing 0.1M NaBH$_4$ and 1M NaOH. After completion of reaction, the solution was washed with de-ionized water, filtered and dried by vacuum filtration using a filter. The recovered Pt loaded MWNC were dried at 80°C for 3 h. For synthesis of Pt-Ru/MWNC, same procedure discussed above was used by taking both precursors 0.075 M H$_2$PtCl$_6$ and 0.15 M RuCl$_2$ solutions.

![Fig. S2: (A) TEM and (B) HRTEM images of Pt/MWNC.](image-url)
Fig. S3: Powder X-ray diffractograms of Pt/MWNC.

S4: Fabrication of PEMFC and DMFC:

For the fabrication of PEM fuel cell, the membrane electrode assembly (MEA) was obtained by sandwiching a pre-treated Nafion membrane (Nafion®1135 and Nafion®1110) between the anode and the cathode. Nafion membranes cut in the required dimension was pretreated by boiling in a solution of 5% H₂O₂ and 1 M H₂SO₄ at 80°C for 30 min and washing with deionized water, and placed on a vacuum table preheated to 80°C. The vacuum table was used to hold the membrane in place and avoid wrinkling during the catalyst application. Both the anode and cathode layers consisted of a backing layer, a gas diffusion layer and a catalyst layer.

Carbon black (Vulcan Xc-72) was treated at 600°C for 3 h to remove organic matter. Next, the pretreated carbon was mixed with polytetrafluoroethylene (PTFE) and isopropyl
alcohol in a supersonic mixer. Finally, this mixture was applied on wet-proofed carbon cloth and dried at 300°C. To form a hydrophilic layer, glycerol was added to the mixture and sprayed on the electrode. The uncatalyzed carbon electrode was cut to a proper size and installed in a sample holder for coating catalyst layer. To prepare the catalyst layer, the required amount of catalyst was suspended in de-ionized water and ultrasonicated by adding 5 wt. % Nafion solution. The amount of Nafion loading was controlled to 1.2 mg/cm². The suspension was spread uniformly over a carbon electrode by spin coating technique. MEA was prepared by sandwiching the Nafion membrane between two electrodes by hot pressing at 130 ºC and 70bar for 2 min. For PEMFC, the anode was Pt/MWNC electrode (3.4 x 3.4 cm² in area and Pt content of 20 wt.%), with a platinum loading of 0.25 mg/cm² and the cathode was prepared from a suspension containing mixture of Pt/MWNCs (Pt content of 20 wt %), with a platinum loading of 0.5 mg/cm². For DMFC, anode was Pt–Ru/MWNC, with a loading of 2.5 mg/cm² and the cathode was Pt/MWNC, with a platinum loading of 2.5 mg/cm².

A single PEMFC and DMFC was assembled using the MEA, two graphite plates with gas channels machined with a serpentine geometry, two Teflon gaskets and two aluminum end plates. The performance of the cells was studied in an indigenously fabricated Fuel Cell test station, using a dc electronic load box. The performance of PEMFC was carried out under an operating pressure of 1 bar of oxygen and hydrogen and for DMFC, 1 M methanol was passed at a rate of 1ml/min. by maintaining oxygen operating pressure of 1 bar.

**S5: Determination of Hydrogen storage capacity:**

Hydrogen adsorption studies have been carried out for the as-grown, purified MWNC and Pt dispersed MWNC in the pressure range 1-100 bar and at 298 K, using a high pressure Seiverts’ apparatus, wherein the necessary precautions were taken during the adsorption measurements. The low sample density of MWNC was also taken into account while calculating the adsorption capacity. About 0.2 g of the sample is loaded in the sample cell and
evacuated to 10^{-6} Torr after flushing with hydrogen 2-3 times. Activation of the samples were done by heating it at 120°C for about 4 hours and the hydrogen at a particular pressure is allowed to react with the sample and cooled to the desired temperature. The equilibrium pressure drop was observed for 2 h and was noted down. The amount of hydrogen adsorbed was calculated from the drop in pressure. After each cycle the sample was degassed for 3 h at 300°C. Prior to hydrogen storage measurements, experiments were conducted to make sure that the system was leak free. The hydrogen adsorption capacity of MWNC is expressed in terms of wt. % (\frac{\text{weight of the hydrogen molecules}}{\text{weight of the material}} \times 100). In the present work, the hydrogen storage capacity was determined by gaseometric technique in which the hydrogen concentration present in the material was obtained from the pressure reduction technique. In this technique, the number of moles of hydrogen atoms adsorbed per mole of the sample is calculated from the pressure change observed before and after hydrogen adsorption at a constant calibrated volume and at a constant temperature. At higher hydrogen gas pressures, the ideal gas law was corrected using the van der Waals equation for the volume of gas molecules and molecular interactions.

**S6: Construction of Supercapacitor:**

The electrodes were pellets (12 mm diameter and 0.1 mm thickness) prepared by pressing the functionalized MWNCs using a binder (PVDF) at 500 kg/cm² and later pellets were heated at a temperature of 120°C in vacuum to remove the binder. Two symmetric electrodes were sandwiched in a Swagelok type stainless steel (SS) test cell with a thin polymer separator (Celgard® 3400) using gold coated SS current collectors. The system was kept under pressure of 16 kg/cm² using Swagelock. The electrochemical properties and capacitance measurements of these electrodes were studied using a two-electrode system with 1M H₂SO₄ aqueous solution as electrolyte. Cyclic voltammetry and galvanostatic charge-discharge measurements have been carried out using an electrochemical workstation PSGSTAT-30 (AUTOLAB).
Voltammetry testing was carried out at potentials between 0 V and 1.0 V and charge-discharge was carried with constant current of 10 mA in the potential range between 0 and 1.0 V using 1 M H\textsubscript{2}SO\textsubscript{4} aqueous electrolyte solution. Since the measurements are made on symmetric assemblies of MWNC, by the basic circuit relationship for series capacitors what is measured is actually $\frac{1}{2}$ of the capacitance of the freestanding electrode.