# **Preparation and Characterization of Hybrid Conducting Polymer – Carbon Nanotube Yarn**

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# **Supporting Information:**

## **Preparation of PPy-CNT Yarn**

CNT yarns (1-ply and 2-ply) were used to produce the PPy-CNT nanocomposite yarn. PPy-CNT yarns were developed by chemical polymerization of pyrrole through vapour phase polymerization. A length of CNT yarn was fixed into a frame (Fig. S1-a) which was dipped into Fe.*p*TS (oxidant/dopant) solution for 10 min (Fig. S1-b). The CNT yarn coated with Fe.*p*TS was then dried at 60°C for 30 min (Fig. S1-c). The dried CNT/Fe.*p*TS yarn was placed in a chamber containing monomer (pyrrole) for 12 hr at room temperature (Fig. S1-d). Polypyrrole then formed on and within the CNT yarn (Fig. S1-e). The PPy-CNT yarn was washed with methanol to remove salts and/ or monomer from the sample (Fig. S1-f). As-prepared PPy-CNT yarn was dried at room temperature and used for further experiments. A comparison of the weight of sample before and after polymerization indicated that the weight fraction of the PPy in the PPy-CNT yarn was ~ 8 wt%.

### **Electrochemical Preparation of PPy-CNT Yarn**

Polypyrrole was incorporated into the CNT yarn successfully by anodic oxidation of pyrrole monomer. CNT yarns (2 ply) were attached to a frame which was used as the anode and a stainless steel plate as the cathode (Figure S2-a). The electrolytic cell contained 0.10 M Fe.pTS, 0.10 M pyrrole and water (Figure S2). The polymerization was carried out galvanostatically using a constant current of 0.10 mA/cm<sup>2</sup> for 6 hr at -0°C. The geometric surface area of a 20 µm diameter

yarn was used to estimate the surface area. The resultant yarn was washed several times with acetone, and then allowed to dry for 24 hr in air at room temperature (Figure S2-b). A comparison of the weight of sample before and after polymerization indicated that the weight fraction of PPy in the electrochemical prepared PPy-CNT yarn has  $\sim$  74 wt%.



**Figure S1:** Preparation of PPy/CNT yarn using vapour phase polymerization; (a) pristine CNT yarn (b) CNT yarn into oxidant/dopant solution (c) Drying (d) Polymerization (e) as-prepared CNT/PPy yarn (f) Washing



**Figure S2:** Preparation of PPy-CNT yarn using electropolymerization; (a) pristine CNT yarn and electrochemical cell; (b) photograph of yarn after polymerization showing the coated yarn and a portion of uncoated yarn that was above the electrolyte solution.

### **Raman Spectra of PPy-CNT Yarn**

Raman spectroscopy was used to investigate the composite structure of the PPy-CNT yarns (Figures S3(a-c)). The spectra show multiple, broad peaks that approximately correspond to those arising from PPy. The spectra are very similar to those given in a previous report for electrochemically prepared PPy[1]. The results can be taken as evidence that doped PPy had been successfully coated on/ infiltrated into the CNT yarn. The important features of the Raman spectra for multi walled carbon nanotubes occur in the range 1330–1590 cm<sup>-1</sup>. A band is seen at 1330 cm<sup>-1</sup> (D band) in Figure S3(a), which has been attributed to the breakdown of translational symmetry produced by the microcrystalline structure [2]. A peak is seen at 1590 cm<sup>-1</sup> that corresponds to one of the  $E_{2g}$  modes [3]. The Raman band at 2660 cm<sup>-1</sup> (G' band) is the overtone of the D band (Figure S3(a)). In addition, Figures S3(b-c) show the Raman spectra of chemically polymerized PPy-CNT yarn, and an electropolymerized PPy-CNT yarn, respectively. Major Raman peaks for PPy occur at 934, 969, 1053, 1076 and 1581 cm<sup>-1</sup> [4, 5]. The spectra shown in Figures S3(b-c) show multiple, broad peaks that approximately correspond to those arising from PPy. The spectra are very similar to that given for electrochemically prepared PPy-DEHS [1]. The results can be taken as evidence that doped PPy had been successfully coated on/ infiltrated into the CNT yarn.



**Figure S3:** Raman spectra of (a) the pristine MWNT yarn, (b) chemically and (c) electrochemically modified the PPy-CNT yarn.



**Figure** S4:Ln  $\sigma$  vs. T-1/(1+d) (a) d=1 (1D-VRH), (b) d=2 (2D-VRH), and (c)d=3 (3D-VRH) of the pristine CNT yarn and the electrochemically and chemically prepared CNT-PPy yarns.



**Figure S5.** The magnetoresistance as a function of B at different temperatures for (a) CNT yarn, (b) Electrochemically prepared CNT-PPy yarn, and (c) Chemically prepared CNT-PPy yarn.



**Figure S6.** Magnetoresistance *vs.*  $B^{1/2}$  for (a) CNT yarn, (b) Electrochemically prepared CNT-PPy yarn, and (c) Chemically prepared CNT-PPy yarn. Lines are shown to act as a guide. The arrow shows the crossover field  $B^*$ .

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