

**Supporting information for:**

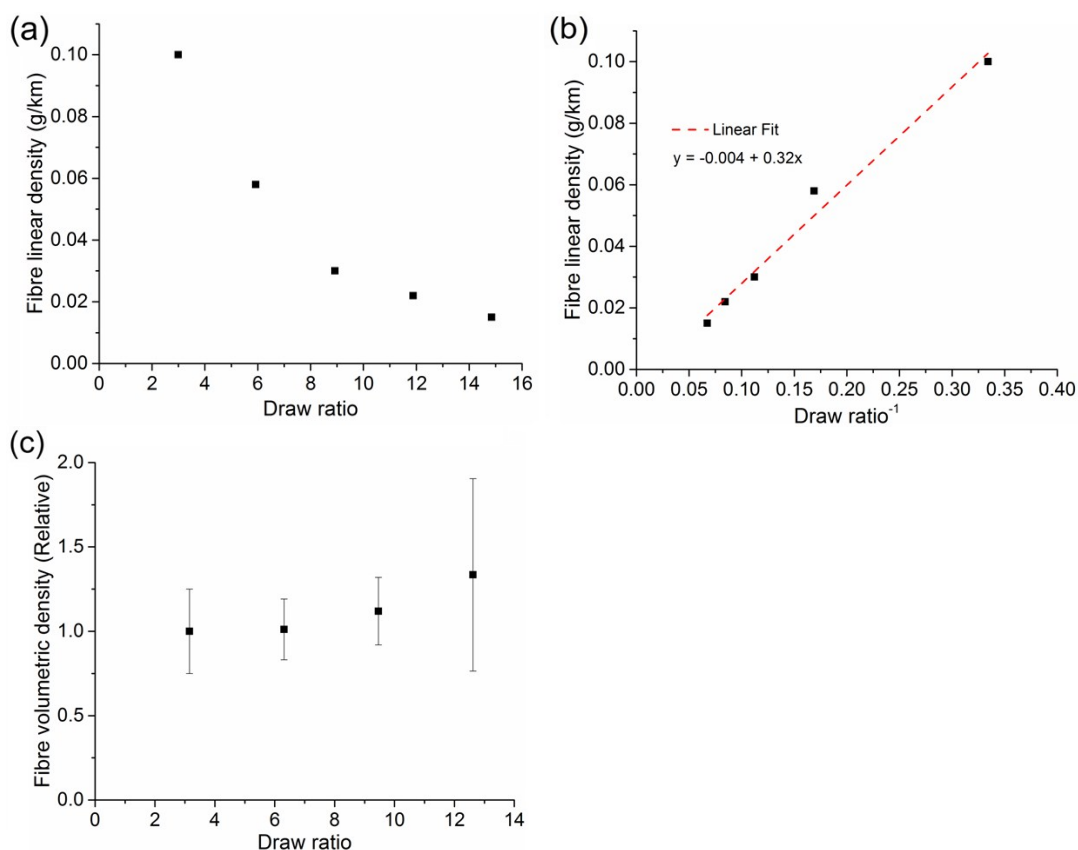
**Macroscopic Fibres of CNTs as Electrodes for Multifunctional Electric  
Double Layer Capacitors: from Quantum Capacitance to Device  
Performance**

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## 1. The effect of draw ratio on fibre linear and volumetric density.

Figure 1 presents the plots of fibre linear and volumetric densities against draw ratio. The linear density decreases with increasing winding rate following the expected inverse relation. As the winding rate approaches the gas-flow rate in the reactor, the determination of the exact minimum spinning rate is not very precise. Thus the draw ratio of 1.5 is not taken into account for the fitting. Volumetric fibre density increases at higher winding rates as the higher degree of alignment improves packing.

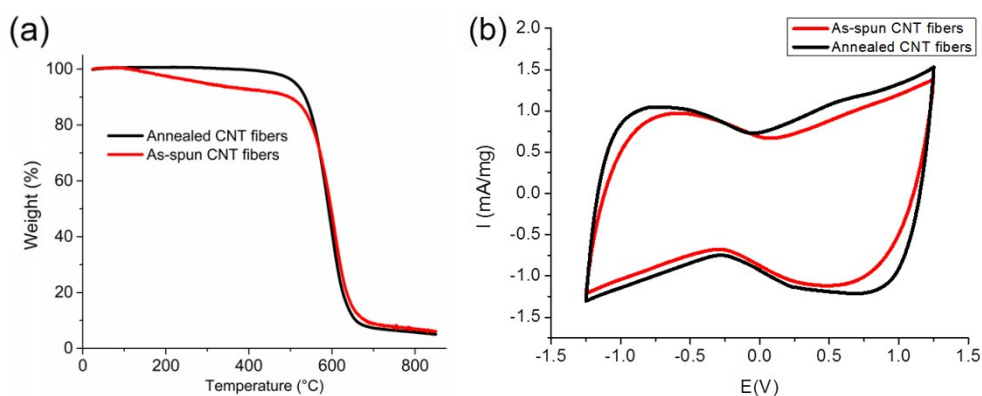


**Fig. S1.** (a) Influence of draw ratio on the fibre linear density b) showing linear dependence as expected and c) dependence of fibre volumetric density on the draw ratio.

## 2. Electrochemical measurements on annealed fibres

To ensure the electrochemical properties determined are not substantially affected by the presence of impurities, we have carried out selected experiments on samples subjected to an annealing process.

Figure S1a presents a typical curve obtained from thermogravimetric analysis showing the presence of around 10wt.% organic impurities produced at the end of the CVD reactor and which adsorb on the surface of the as-spun CNT fibers. These TGA weight losses below 400 °C, are attributed to desorption of water and oxidative decomposition of carbon impurities, as determined by infrared spectroscopy.<sup>1</sup> These impurities have been removed from the samples by annealing the fibers in argon atmosphere at 400 °C for 2 hours, as confirmed by TGA (Figure S2a (black line)). These higher purity sample shows nearly identical CV compared to the as-spun material, including the so-called butterfly shape due to the quantum capacitance of the system (Figure S2b).



**Fig. S2.** (a) TG curves (in N<sub>2</sub>) of as-spun and annealed CNT fibers samples. (b) CV curves of as-spun and annealed CNT fibers at scan rate of 50 mV/s and 2.5V of voltage window.

1. Sundaram, R. M. Production , Characterisation and Properties of Carbon Nanotube fibres. (2011).