Supplementary

## Carbon nano-fiber forest foundation for ruthenium oxide pseudo-electrochemical capacitors

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**SFig.1** Electron microscopy images of the  $RuO_2$ -CNF electrode prepared at 250 °C (a) BSE, (b) SE, and (c) SE image at a higher magnification showing a spot with open structures even with the oxide coating on it. (d) Energy dispersive spectra (EDX) obtained from the white box shown in image (c).



**SFig. 2** EDX maps showing the distribution of elements present in the RuO<sub>2</sub>-CNF electrode prepared at 250 °C along with its corresponding secondary electron image.



SFig. 3 TEM image of uncoated CNF. No hollow core can be observed in these CNF.



**SFig. 4** (a) Raman spectra of the uncoated CNF (in red) and the RuO<sub>2</sub>-coated CNF (in black) prepared at 250 °C. Peak positions were determined by fitting the curves with Lorentzian functions, after background subtraction and data normalization. Enlarged Raman spectra of (b) RuO<sub>2</sub>-coated

CNF showing poorly pronounced peaks at 520 cm<sup>-1</sup> and 640 cm<sup>-1</sup>. (c) CNF showing no such peaks.

**STable 1** Raman characterisation of CNF forest to calculate the crystallite size (La) and the defect density  $(I_D/I_G)$ .

Fit parameters for D and G bands using Lorentzian function		
D Peak, cm <sup>-1</sup>	$1344.43 \pm 0.86$	
G peak, cm <sup>-1</sup>	$1586.34 \pm 0.92$	
D band FWHM, cm <sup>-1</sup>	$151.52\pm6.90$	
G band FWHM, cm <sup>-1</sup>	$79.56\pm0.92$	
$I_D/I_G$	$2.28\pm0.08$	
La (nm)	$2.21\pm0.08$	

 $L_a$  is calculated using Equation 1<sup>-1</sup>. Where  $L_a$  in nm and  $E_L$  is the excitation energy in eV In this study EL is 2.33 eV corresponding to the 532 nm laser.

$$L_a = \frac{4 \cdot 4}{\frac{I_D}{I_G}} \left(\frac{2.41}{E_L}\right)^4 \tag{1}$$



**SFig. 5** (a) TEM showing  $RuO_2$  clusters adhered to CNF.(b) HR-TEM of a  $RuO_2$ -coated CNF prepared at 250 °C. (c) EDX spectra performed within the region shown in a white square in SFig. 4a. It is to be noted that the imaged specimen was taken from the electrode and mounted on to the TEM grids for imaging; therefore, the images do not represent the true electrode structure when originally formed on the Ni-f substrate.

Compound	Peak BE (eV)	FWHM (eV)
O1s (RuO <sub>2</sub> )	$530.35\pm0.09$	$2.20\pm0.08$
O1s (C=O)	$532.43 \pm 0.15$	$3.35\pm0.18$
$Ru_3p_3$ ( $RuO_2$ )	$462.87\pm0.22$	$3.49\pm0.01$
$Ru_3p_1$ ( $RuO_2$ )	$484.96 \pm 0.15$	$3.49\pm0.01$
Ru <sub>3</sub> p <sub>3</sub> (RuCl <sub>3</sub> )	$466.02\pm0.30$	$3.48\pm0.02$
$Ru_3p_1$ ( $RuCl_3$ )	$488.07\pm0.20$	$3.48\pm0.02$
$Cl_2p_3$ (Ru $Cl_3$ )	$198.29\pm0.02$	$2.35\pm0.24$
$Cl_2p_1$ (Ru $Cl_3$ )	$199.99 \pm 0.02$	$2.35 \pm 0.24$

STable 2 Fitted peak values for all the HR-XPS discussed in the main article



**SFig. 6** TGA curves for (a) CNF on Ni-f (in black) and RuO<sub>2</sub>-CNF on Ni-f prepared at 250 °C (in red), (b) hydrous RuCl<sub>3</sub> (reproduced from ref. 2, Copyright © 2018 Sebastian Chalupczok et al.)



**SFig. 7** (a) CVs of a RuO<sub>2</sub>-CNF electrode prepared at 200 °C, recorded at scan rates of 5, 10, 20, 50, 80, 100, 200 mV/s (in the direction of the arrows, respectively). (b) CV recorded at 5 mV/s showing broad anodic and cathodic peaks. (c) Peak currents as a function of scan rate showing a linear relation. Red-dotted lines are the linear fit, with the equation of  $y=4.70\times10^{-4} x$  and  $y=4.58\times10^{-4} x$  for the anodic and the cathodic current data, respectively.



SFig. 8 Mass accumulation of RuO<sub>2</sub> on CNF directly grown on Ni-f.



**SFig. 9** Comparison of Nyquist plots for  $RuO_2$ -CNF electrodes prepared at 250 °C recorded in a three- and two-electrode cell. Three-electrode-cell measurement show higher series resistance than the two-electrode-cell measurement.

With a series RC model to fit the EIS data, the real C'( $\omega$ ) and imaginary C"( $\omega$ ) parts of capacitances were calculated using Equation 2 and Equation 3<sup>3</sup>.

$$C'(\omega) = \frac{-Z''}{\omega |Z(\omega)|^2}$$
 Equation 2

$$C''(\omega) = \frac{Z'}{\omega |Z(\omega)|^2}$$
 Equation 3

Where, -Z" and Z' are imaginary and real components of the impedance (Z, ohms),  $\omega = 2\pi f$ , and f is the frequency (Hz).

The relationship of the capacitance with frequency is shown in SFig. 9 (a and b) for the two and three-electrode measurements respectively. This demonstrates the decrease in C'(w) with an increase in frequency as observed previously in the Nyquist plot. C"(w) indicates the energy dissipation and this increases with the increase in frequency (note: capacitances are reported per electrode). As the peak C" shifts to the higher frequency in the two-electrode cell than the three-electrode cell (SFig. 9), it is evident that the delivery of stored energy is slightly faster (i.e. delivers energy in less time) in the later cell setup at higher power.



**SFig. 10** X-ray diffraction pattern of RuO<sub>2</sub>-CNF electrode surface prepared at 250 °C. The strong occurrence of nickel peaks makes it hard to observe peaks of RuO<sub>2</sub>. However, a small peak at 2 $\theta$  = 26 can be ascribed to the carbon fibers <sup>4</sup>. The peaks at 2 $\theta$ ; 44.45, 52.05, 76.62, 92.62, 98.16 all correspond to nickel. X-ray diffraction paters collected using Cu K-alpha X-ray source (D8 Discover Bruker).

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