## Electronic Supplementary Information: Reductive Dissolution of Supergrowth Single-Walled Carbon Nanotubes for Tougher Nanocomposites

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# 1. Supplementary scanning electron micrographs



**Figure S1.** SEM of as-received SG SWCNT forests, illustrating in red SWCNT length in micrometres, scale bars represent 200  $\mu$ m in all images.



Figure S2. High magnification SEM of filtered NaNp/DMAc SG-CNT solution

#### 2. Discussion of Initial SG-CNT Length

The length of SWCNTs within forests from pre-deposited catalyst CVD, particularly for ultra-long SWCNTs, is a contentious point within the CNT community. As SWCNTs do not grow directly perpendicular to the substrate (instead growing in a wavy manner), the forest height is actually marginally smaller than the projected length of SWCNTs, should the nanotubes grow continuously, although the differences are typically on the order of 10-20% (E. Meshot et al., Nanoscale, 2010, 2, 896-900). However, direct microscopic observation of individual long SWCNTs spanning the entire forest height have not been reported, and the assumption of continuous SWCNTs along the forest height has to date primarily relied on circumstantial evidence. For example, random activation and deactivation of catalysts to produce forests would lead to variations in forest height, density, and hierarchical order across the forest, due to the non-linear growth kinetics of CNTs (Einarsson et al., Carbon, 2008, 48, 923-930) as well as the observation of CNT ends. The diameter distribution from radial breathing modes in Raman line mapping along a forest is also known to be consistent (Futaba et al., J. Phys. Chem. B, 2006, 110, 8035-8038) consistent with vertical uniformity. Additionally, deliberately interrupted growth leads to a uniform banding structure in SEM micrographs (Dijon et al., Carbon, 2010, 3953-3963 and Zhang et al., J. Phys. Chem. C, 2010, 114, 6389-6395).

In this work, the SEMs of as-received material (Main text, Fig. 1-2, ESI Fig. S1) consistently show SG-CNT forests with even widths, even colouration along the forest height, and no obvious CNT ends in the middle of the forest (Main text, Fig. 2, top). While not indisputable, the evidence is consistent with the constituent CNTs spanning the forest, as is typically assumed within the field.

## 3. Concentration of reduced SG-CNT solutions.



**Figure S3.** Concentration (and yield) of 1 mg mL-1 SG-CNTs left to stir for 2 weeks as a function of charging stoichiometry post centrifugation (1,000 g, 30 min)



**Figure S4.** AFM of NaNp/DMAc dispersed SG SWCNTs with height profiles for each line indicated adjacently. The greyed regions indicate peaks which were not attributable to SWCNTs, also indicated on the AFM.

# 4. Supplementary atomic force microscopy



**Figure S5.** AFM of SG SWCNTs dispersed through sonication in NMP with height profiles for each line indicated adjacently.



**Figure S6.** AFM of SG SWCNTs dispersed through sonication in aqueous SDS solution with height profiles for each line indicated adjacently.



**Figure S7.** AFM micrograph of purified Elicarb SWCNTs showing 0.5-1.2  $\mu$ m SWCNTs. Length distribution of the Tuball SWCNTs used here (Lot # 4-18032014, 1.288 ± 1.065  $\mu$ m) can be found in A. J. Clancy *et al.*, Carbon, 2016, 108, 423–432.

## 5. Statistical Raman analysis



**Figure S8.** Statistical D/G mode intensity ratios from Raman spectroscopy of SWCNTs used for composite fibre dopes.

## 6. Supplementary thermogravimetric analysis



Figure S9. TGA thermographs of as-received and purified SWCNTs in air.



**Figure S10.** TGA of PVC (44 kDa) in N2; 8 wt.% char is typical of unmodified PVC (J. Yu, L. Sun, C. Ma, Y. Qiao and H. Yao, *Waste Manag.*, Thermal degradation of PVC: A review, **2016**, *48*, 300-314).



**Figure S11.** TGA grafting controls. Tuball SWCNTs and SG-CNTs mixed with 10 wt eq. PVC after both sonication in NMP and reduction with NaNp/DMAc. TGA of SG-CNT and Tuball samples were performed on a Mettler Toledo TGA/DSC1 under N<sub>2</sub> flow at 60 sccm with manually subtracted blank-run backgrounds on 0.5-1 mg of sample. Samples were held at 100°C for 30 min before heating at 10 °C min<sup>-1</sup> to 700 °C.

7. Additional SEM PVC/SWCNT Nanocomposite Fibres



**Figure S12.** SEM of PVC/CNT Elicarb nanocomposite fibres. (a) Fracture surface of unwashed Elicarb/PVC fibre showing dewetted bulk PVC along bundles. (b) Tuball/PVC composite fibre surface before soaking fibre in warm MiBK, (c) Tuball/PVC composite fibre surface after soaking in MiBK, (d) Low magnification of post-soaked Tuball/PVC composite fibre.



## 8. Coagulation spinning equipment

Figure S12. Schematic (top) and picture (bottom) of coagulation spinning rig.

Comula	Wt.%	Wt.%	σ (MPa)		E <sub>Y</sub> (GPa)		ε <sub>f</sub> (%)		U <sub>τ</sub> (MJ m <sup>-3</sup> )		D <sub>f</sub> (μm)	S value	
Sample	<i>ლ</i> 550 ℃	SWCNT	Value (± SE)	SD	Value (± SE)	SD	Value (± SE)	SD	Value (± SE)	SD	(± SD)	As-spun (± SD)	Post- tensile (SD)
Elicarb	32.9	27.1	146 (6.1)	86.6	8.74 (0.03)	0.10	3.89 (0.35)	1.49	3.59 (0.33)	2.36	54.4 (14.7)	0.151 (0.080)	0.251 (0.043)
Tuball	22.8	16.1	203 (14.4)	62.6	7.59 (0.02)	0.09	9.31 (2.13)	2.54	12.7 (2.92)	4.29	53 (8.24)	0.127 (0.053)	0.134 (0.038)
SG-CNT	29.1	22.9	185 (12.7)	47.6	9.33 (0.03)	0.10	20.8 (1.26)	4.73	30.8 (1.26)	1.52	42.3 (8.39)	0.104 (0.021)	0.294 (0.035)
Sol. Cast PVC	8.0	0	20.2 (2.1)	6.6	0.27 (0.006)	0.02	190.6 (21.6)	68.3	27.1 (4.76)	15.0	Rectangle 75 x 2000	-	-

#### 9. Tensile, thermogravimetric and polarised Raman tabulated data

**Table S1.** Properties of SWCNT/PVC composite fibres. Weight percentages of SWCNTs were taken from residual weight at 550 °C in N<sub>2</sub> TGAs using Equation S1. Ultimate tensile strength ( $\sigma_u$ ), Young's Modulus ( $E_Y$ ) and strain to failure ( $\varepsilon_f$ ) taken from tensile testing, diameter ( $D_f$ ) taken from optical microscopy. S alignment values measured from polarised Raman spectroscopy. Standard deviation (SD) and standard errors (SE) are given where appropriate.

$$W_{CNT} = 100 - \left(\frac{8}{92} + 1\right) L_{550C}$$

**Equation S1.** Calculation of SWCNT weight percentage (WCNT) from weight loss at 550 °C ( $L_{550C}$ ) accounting for 8 wt.% of PVC forming char during degradation taken from Figure S10.

### 10. SWCNT/PVC film production and Dielectric test



**Figure S13.** Top) Schematic of SG-CNT/PVC dielectric film fabrication, i) PVC/SG-CNT composite fibre is wound around a glass slide, ii) minimal 5 wt.% PVC/MiBK solution is dropcast onto the wound SG SWCNT composite fibres and dried, iii) SG-CNT/PVC composite film is removed from glass substrate, iv) SG-CNT/PVC composite film is cut in half, v) the two halves of the film are then consolidated in 0/90 layup conformation and adhered using warm MiBK. Bottom, digital photographs of wound SG-CNT/PVC composite fibre (left, analogous to the first process step) and final SG-CNT/PVC high-κ dielectric film (right). N.B. PVC control films were produced by solution casting PVC/MiBK solution into a petri dish.

Sample	Thickness (mm)	Capacitance (nF)	Dielectric constant (ε <sub>r</sub> )		
PVC film (control)	0.033	576	2.73		
SG-CNT/PVC film	0.329	197	9.32		

**Table S3.** Dielectric properties of PVC based films, tested on a 1 cm diameter circular cross-section. c = Ct

$$\varepsilon_r = \frac{1}{\varepsilon_0 A}$$

**Equation S2.** dielectric constant of material ( $\varepsilon_r$ ), capacitance in F (C), permittivity of free space in F m<sup>-1</sup> ( $\varepsilon_0$ ), film thickness in mm (t), capacitor cross-sectional area in m<sup>2</sup> (A).

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