

Supplementary material captions

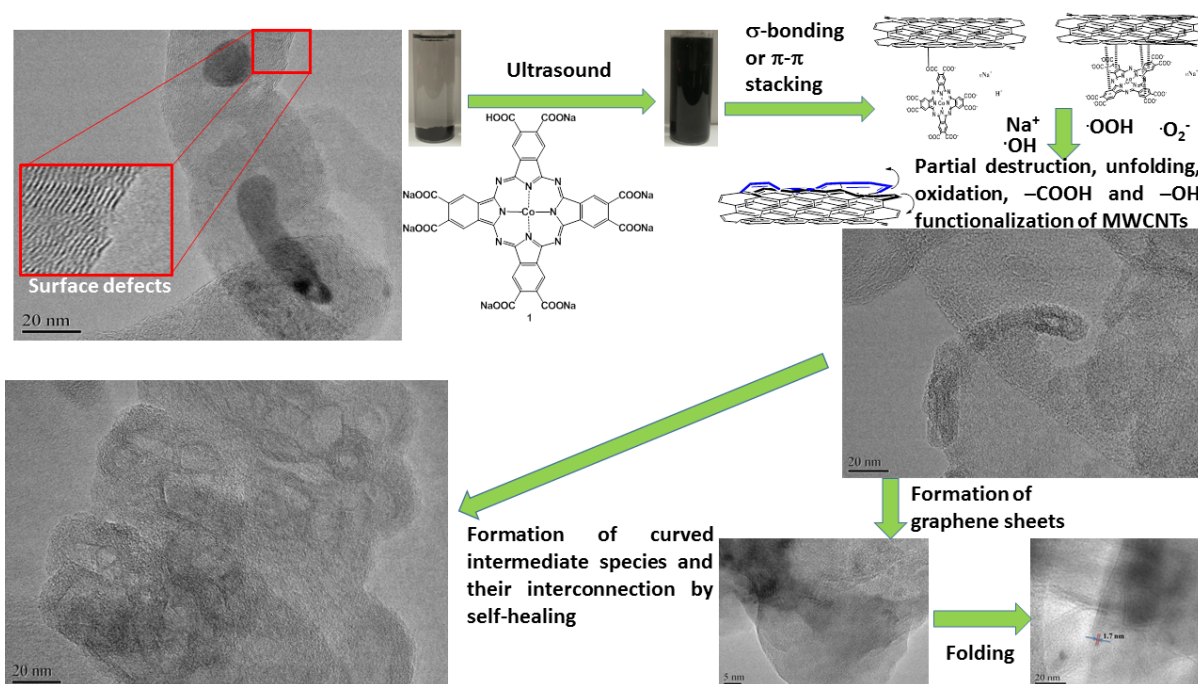
S1. Possible formation mechanism for carbon nano-onions and other products in TP-MWCNTs dispersions.

S2. (Supplementary Table 1). Raman spectra data of MWCNTs-TP interaction products and their comparison with those reported earlier.

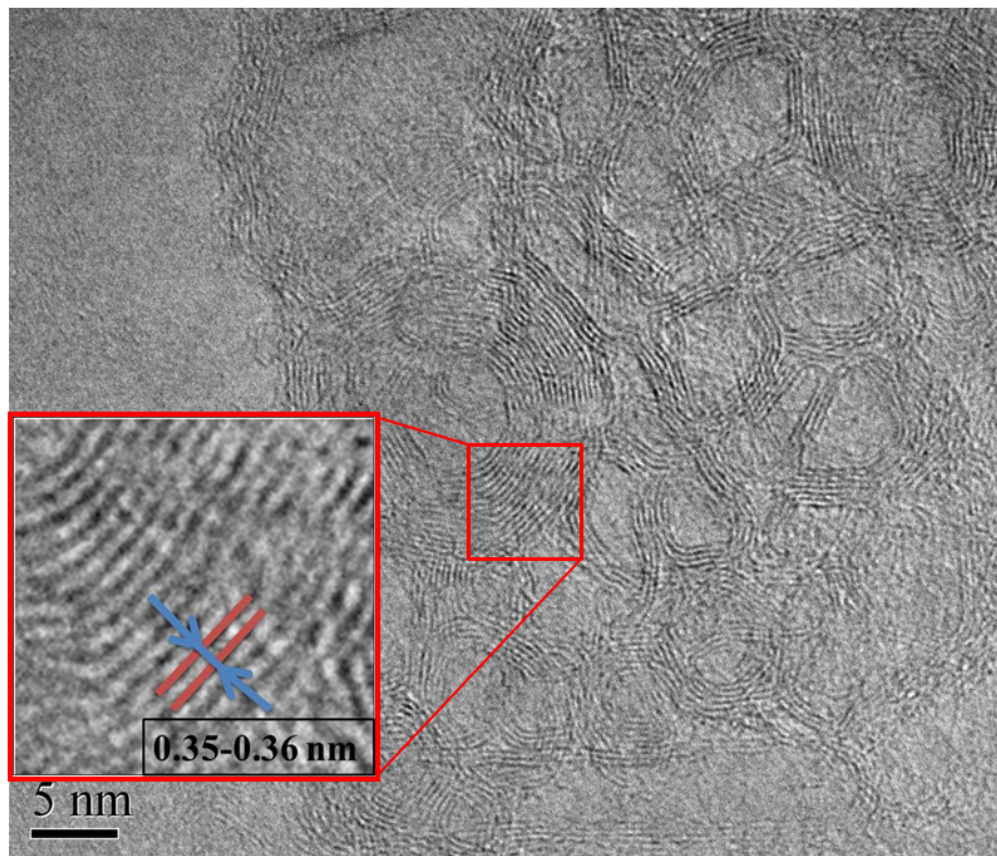
S3. High resolution image of nano-onion structures.

S4. High-resolution image of products in concentrated dispersions TP-MWCNTs.

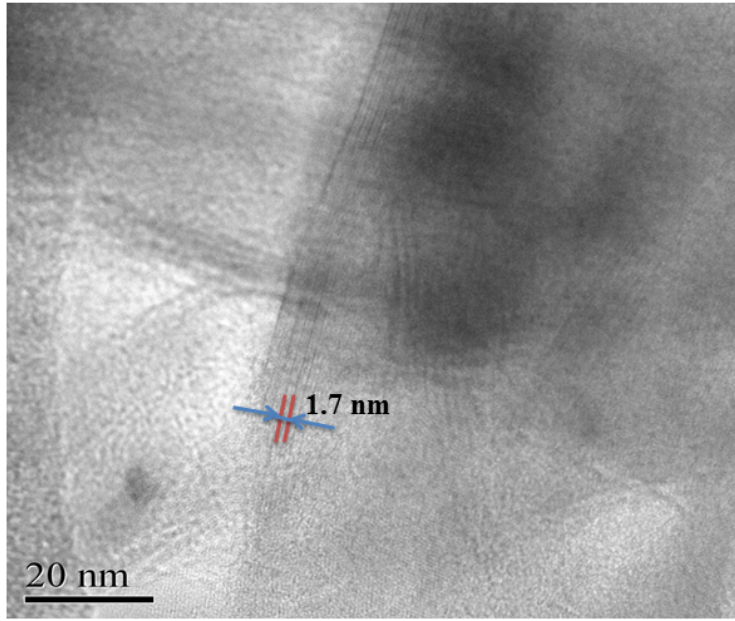
S5. XPS characterization results.



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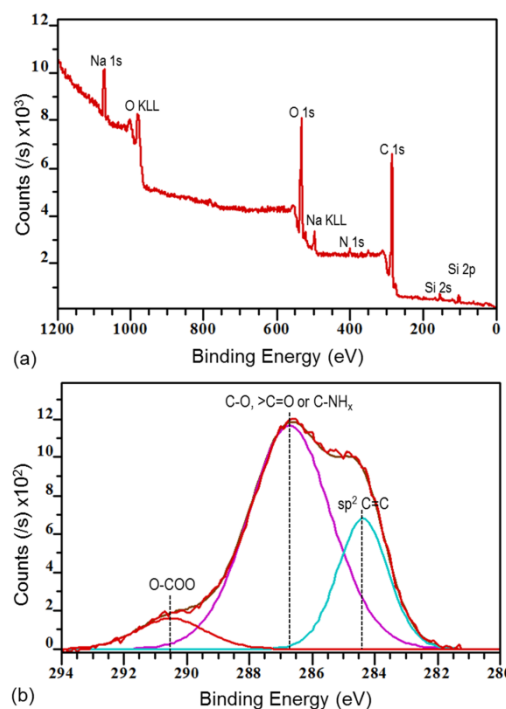
Experimental Details

XPS characterization:

The chemical composition of one selected carbon nanotube sample was investigated by X-ray photoelectron spectroscopy (XPS) on a Perkin-Elmer Phi 560 ESCA/SAM system and the spectra were acquired using a non-monochromated Al $K\alpha$ excitation source. The Al $K\alpha$ excitation is characteristic unresolved $K\alpha_{1,2}$ and has a characteristic energy of 1253.6 eV. Survey scans were conducted in the 0 to 1200-eV range at 0.5-eV steps. High-resolution scans were conducted for C 1s peak in the 280 to 294-eV range at 0.1-eV steps.

Results and Discussion

Fig. (a) presents general XPS survey spectrum of TF-CNT sample exhibiting mainly the presence of C, O, N, Na and Si. This presence of the Si was from the contamination of glass (the samples were synthesized in a glass vial under ultrasonic treatment and dried on glass surface). Fig. (b) shows the high-resolution XPS spectra of the C 1s peak which can be decomposed into the three components at binding energy of 284.4 eV, 286.7 eV and 290.5 eV that can be assigned to sp^2 C=C, C-O (>C=O or C-NH_x) and O-COO, respectively.¹



T.I.T. Okpalugo, P. Papakonstantinou, H. Murphy, J. McLaughlin, N.M.D. Brown, High resolution XPS characterization of chemical functionalized MWCNTs and SWCNTs, Carbon 43 (2005) 153–161

S2. Supplemental Table 1. Raman spectra data of MWCNTs-TP interaction products and their comparison with those reported earlier.

Monomeric TPⁱ		MWCNTs;ⁱⁱ frequency, cm⁻¹	Nano-onions;ⁱⁱⁱ frequency, cm⁻¹	MWCNTs-TP interaction product containing nano-onions; frequency, cm⁻¹
Frequency, cm⁻¹	<i>I</i>_{rel.}, %			
1117	40			1040-1070, 1150
1210	90			
1320	90	1300-1320	1310-1350	1300-1320
1396	40			1390, 1440
1536	100	1590-1600	1600-1630	1580-1610 1530-1540, 1460-1470
		~2700	~2700, ~2800	~2650-2750

ⁱ A.V. Feofanov, A.I. Grichine, L.A. Shitova, T.A. Karmakova, R.I. Yakubovskaya, M. Egret-Charlier, P. Vigny. Confocal Raman Microspectroscopy and Imaging Study of Theraphthal in Living Cancer Cells. *Biophys. J.*, 2000, **78**(1), 499–512.

ⁱⁱ C. Portet, G. Yushin, Y. Gogotsi. Electrochemical performance of carbon onions, nanodiamonds, carbon black and multiwalled nanotubes in electrical double layer capacitors. *Carbon*, 2007, **45**, 2511–2518.

ⁱⁱⁱ J.K. McDonough, A.I. Frolov, V. Presser, J. Niu, C.H. Miller, T. Ubieto, M.V. Fedorov, Y. Gogotsi. Influence of the structure of carbon onions on their electrochemical performance in supercapacitor electrodes. *Carbon*, 2012, **50**, 3298–3309.