

## Supplementary Information

# Morphology – composition correlations in carbon nanotubes synthesised with nitrogen and phosphorus containing precursors

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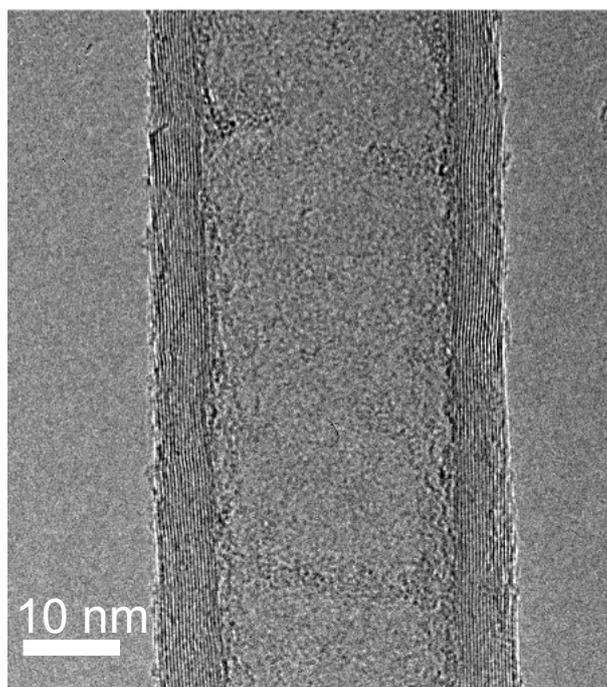
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## Amorphous material on the inner-wall of the nanotube

Amorphous material is shown in the inner channel of the nanotube in Figures 1d, a larger image of this nanotube is shown in Figures S1.



**Figure S1:** Larger version of the nanotube shown in Figure 1d.

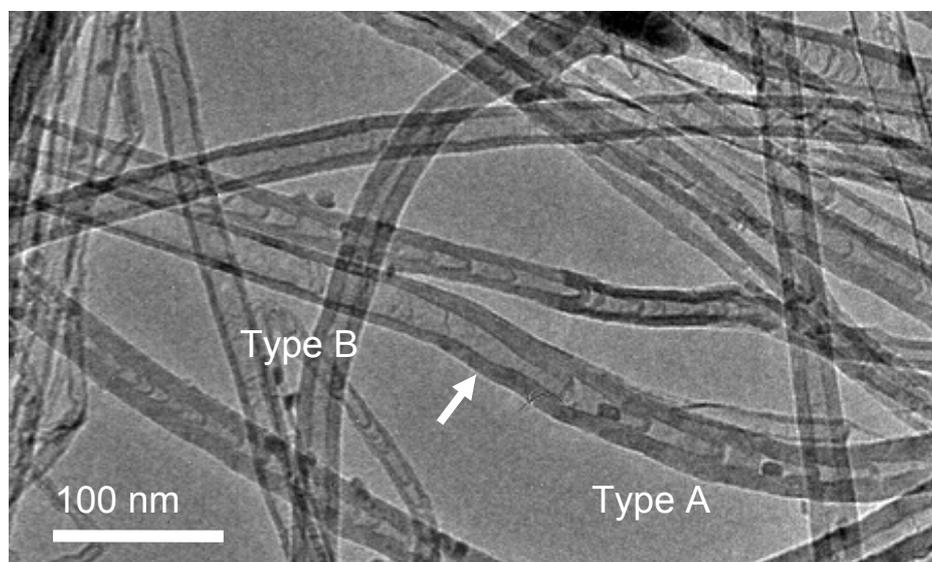
### Proportion of Type A and Type B nanotubes within the sample

The morphology of 216 P,N-MWNTs was studied and a table showing the distribution of the different types of morphology is given below. 77% of the nanotubes, were either Type A or Type B. 20% of the nanotubes were very small, and often curled and containing particles. Although 20% in numbers is a lot, the contribution of these small nanotubes towards the properties of the overall ensemble is expected to be negligible due to their small weight percent. Only 3% of the nanotubes (i.e. 7 out of 216 nanotubes) did not fall into one of these three categories

Morphology	% of sample
Type A	36
Type B	41
Very small nanotubes	20
Other	3

### Transition from a Type A to a Type B nanotube

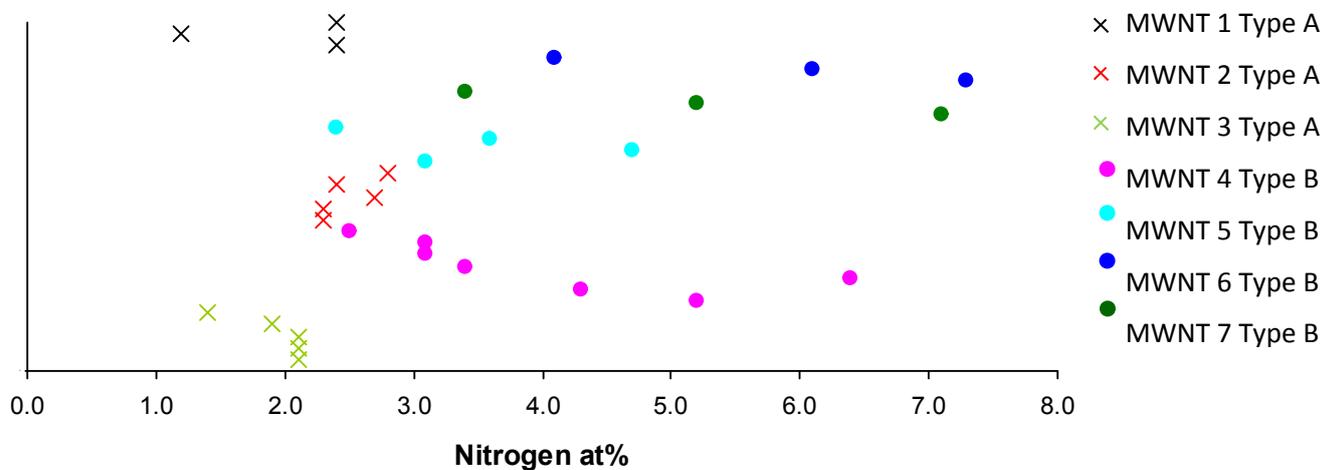
Figure S2 shows a larger image of the nanotubes shown in Figure 1a which changes from Type A (right-hand side) to Type B (left hand side).



**Figure S2:** Nanotube (shown in Figure 1a) which changes from Type A (right-hand side) to Type B (left hand side).

### Variation in nitrogen content along a nanotube

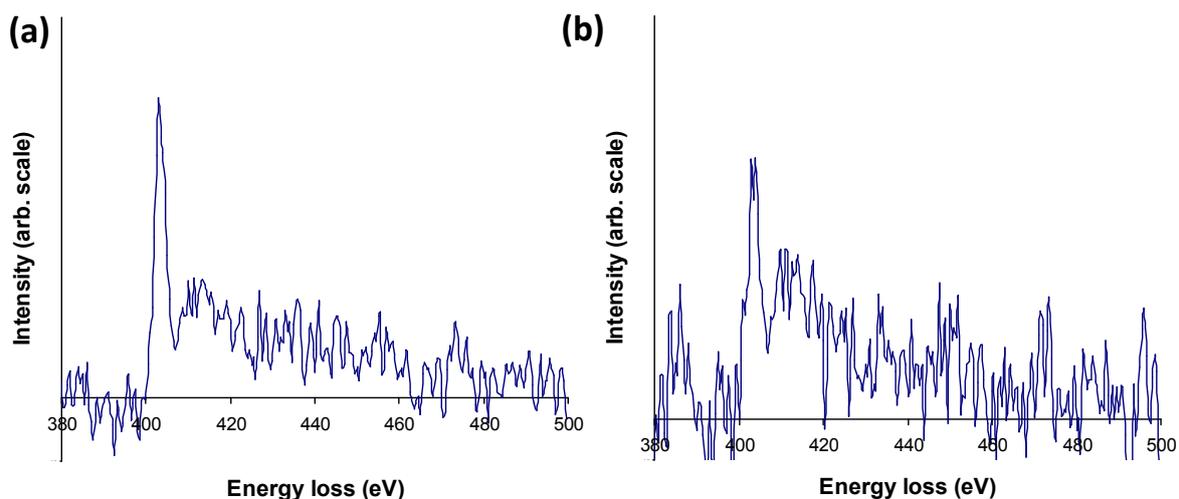
The nitrogen content was measured at different positions in three Type A and four Type B nanotubes (Figure S3).



**Figure S3:** Variation in nitrogen content along seven different nanotubes. Each nanotube is represented by a different colour and each point has been offset by an arbitrary value in the y-direction to make the data easier to interpret.

### Presence of N<sub>2</sub> in the inner channel of a Type B P,N-MWCNT

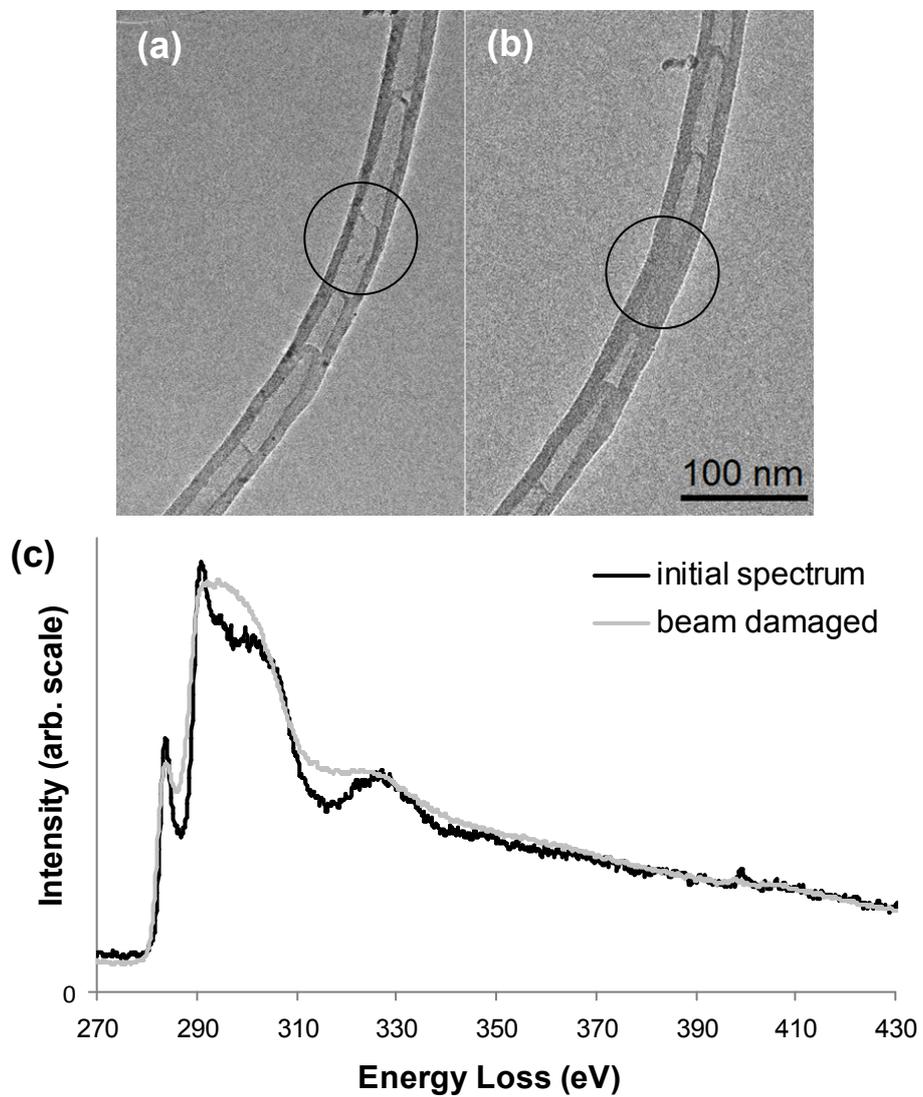
Figure S4a shows the sum of the spectra from pixels 8 – 10 of Figure 3d. The  $\pi^*$  peak is very pronounced, indicating the presence of nitrogen gas. It is likely that the spectrum from an entire Type B nanotube (e.g. Figure 3b) contains a signal from bonded nitrogen on or in the walls of the tube as well as from N<sub>2</sub> gas along the central channel. The combination of bonded nitrogen and N<sub>2</sub> gas will reduce the prominence of the  $\pi^*$  peak compared with only N<sub>2</sub> gas. The nitrogen signal from pixels 7 and 11 does not show such a strong  $\pi^*$  peak and so the total signal from the nanotube (shown in Figure S4b) looks more like that shown in Figure 3b.



**Figure S4:** (a) EELS spectra showing the presence of N<sub>2</sub> gas obtained from the centre of a Type B P,N-MWCNT. (b) The total nitrogen EELS signal from the same Type B P,N-MWCNT.

### Beam irradiation of a Type B P,N-MWCNT

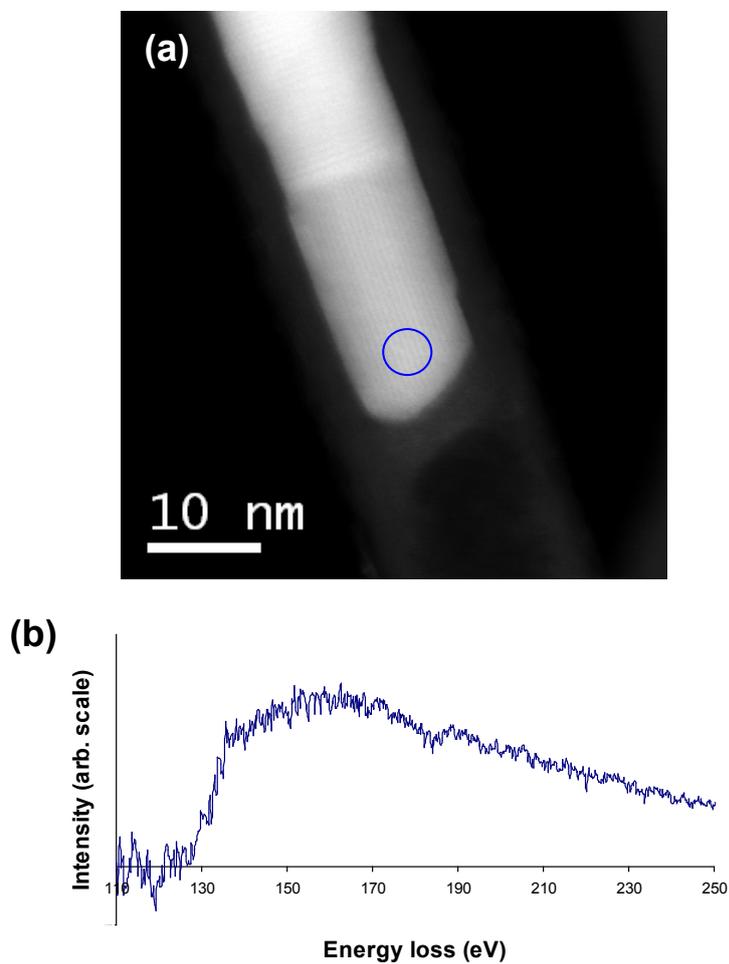
Figure S5b shows the amorphous structure created by beam irradiation of a Type B nanotube. Nitrogen is still detected within the structure, (Figure S5c).



**Figure S5:** TEM micrographs of a nanotube before (a) and after (b) beam irradiation. The corresponding EELS spectra are shown in (c).

### Phosphorus EELS signal from catalyst particle

Figure S6 shows the same catalysis particle as in Figure 4, along with the corresponding phosphorus L-edge EELS signal.



**Figure S6:** HAADF image (a) and phosphorus L-edge (b) from the region marked in (a).