

## Supplementary Information

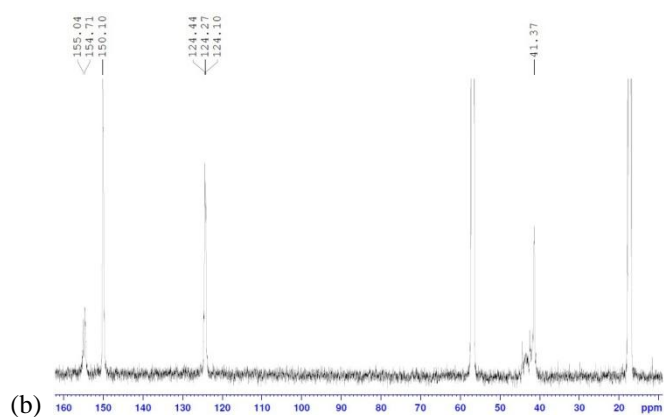
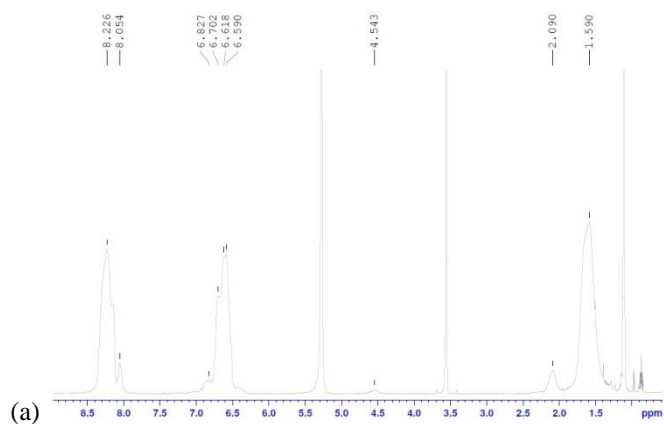
### Supramolecular Approach to Decorate Multi-Walled Carbon Nanotubes with Negatively Charged Iron(II) Complexes

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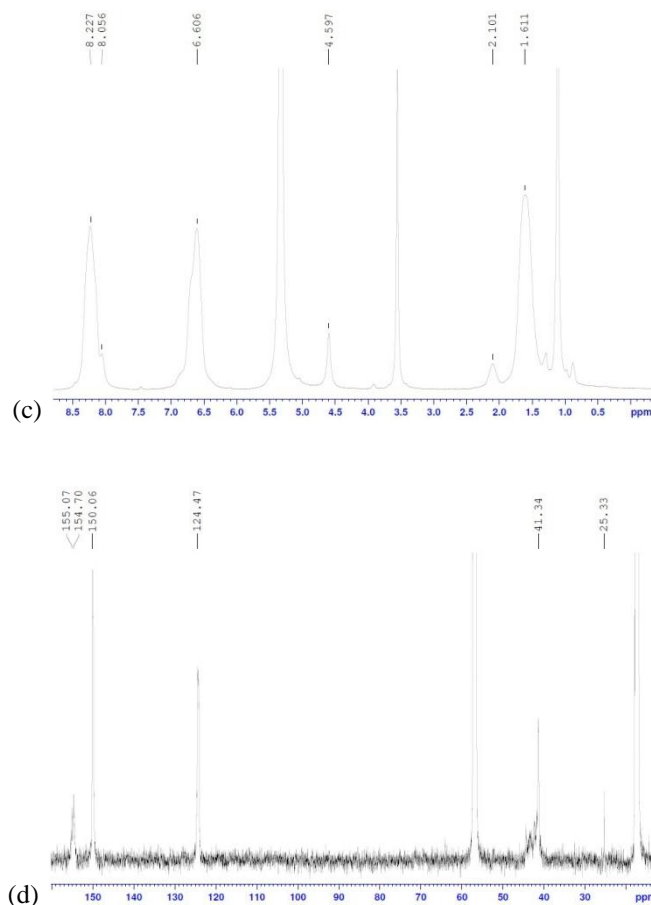
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<sup>1</sup>H and <sup>13</sup>C nuclear magnetic resonance

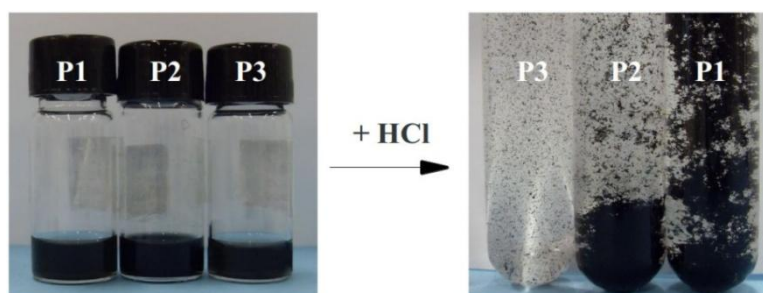


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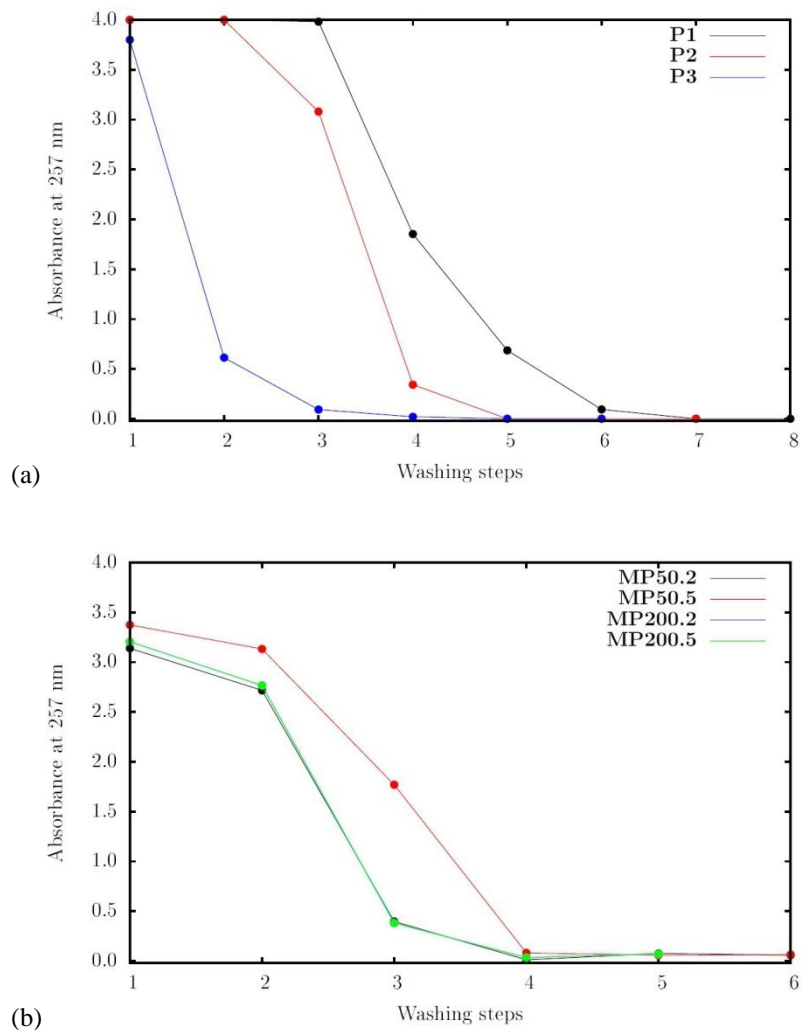
**Figure S1.**  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra (500 MHz, ethanol- $d_6$ ) of P4VP (a,b) and P4VP/MWCNT suspension (c,d).

Determination of the mass of suspended MWCNT



**Figure S2.** Photographs of screw-cap vials containing stable suspensions of the samples and the flocculation induced after addition of concentrated hydrochloric acid.

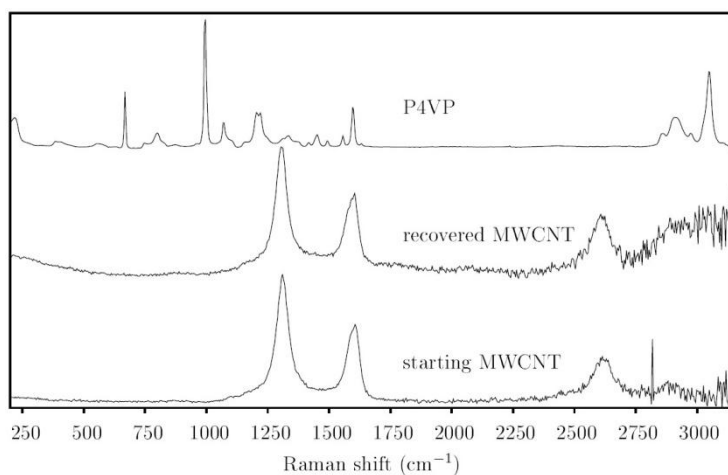
Figure S3 displays the absorbance characteristic of the P4VP as a function of the washing cycles, evidencing that the polymer is removed. Four to seven cycles were necessary to wash P4VP whereas only four cycles were necessary to remove the P4VP- $\text{Fe}(\text{CN})_3^{5-}$  metallopolymer probably due to enhanced hydrophilicity.



**Figure S3.** Plot of absorbance at 257 nm as a function of each washing step with HCl 1.0 mol L<sup>-1</sup> for samples with (a) pure P4VP; (b) with the metallopolymers.

#### Raman spectroscopy of recovered MWCNT

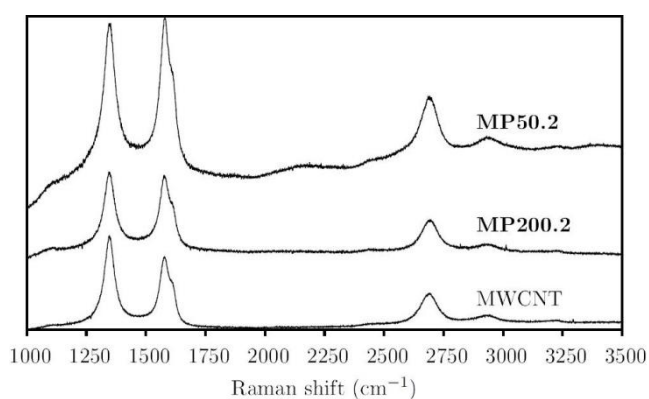
Figure S4 shows the Raman spectra of the starting MWCNT, the P4VP polymer and the MWCNT isolated from the suspension with P4VP through the washing procedure with acid. The absence of bands relative to the polymer indicates that the isolated MWCNT did not contain P4VP, in agreement with the lack of absorption in the UV region (Figure S3).



**Figure S4.** Raman spectra of the starting MWCNT, P4VP and the MWCNT recovered from the acidic treatment of a suspension formed by addition of 17.5 mg of nanotubes in 3.5 mL of ethanol containing 35.0 mg of P4VP.

Fitting details of Raman spectra

Figure S5 shows the non-normalized spectra of Figure 5.



**Figure S5.** Raman spectra of the pristine MWCNT and the composites made with MWCNT and P4VP-Fe(CN)<sub>5</sub> metallopolymer with py:Fe = 50:1 (MP50.2) and py:Fe = 200:1 (MP200.2).

The spectra on Figure S5 were fitted with lorentzian curves of the form:

$$I(\nu) = I_0 + \frac{2A\omega}{\pi^4(\nu - \nu_c)^2 + \omega^2} \quad (S1)$$

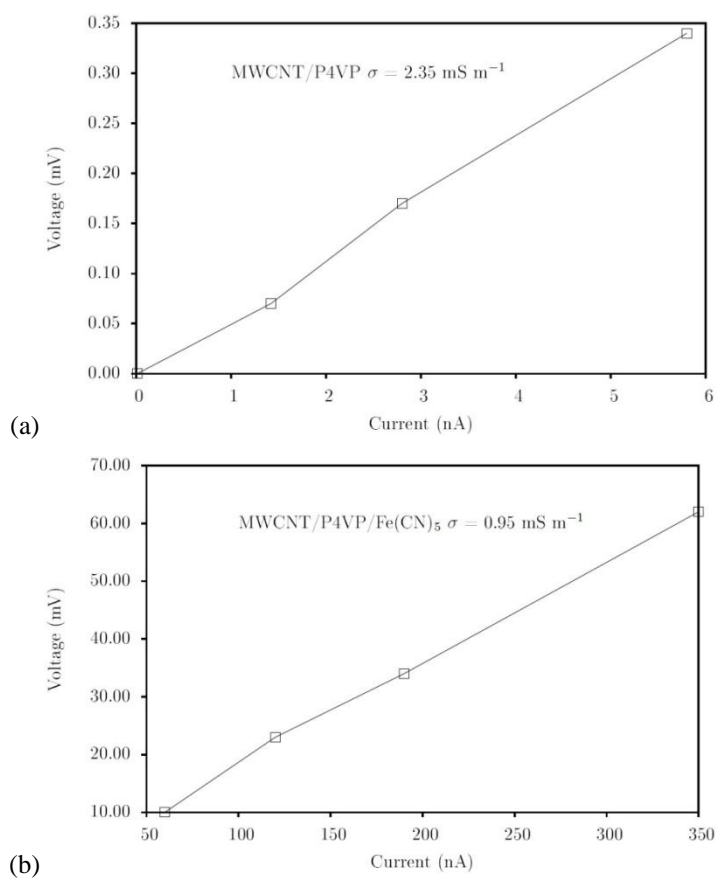
where  $I_0$  is the baseline offset,  $A$  is the area,  $\omega$  is the width and  $\nu_c$  is the wavenumber at maximum intensity. The results of the curve fitting procedure is shown in Table S1.

**Table S1.** Fitting parameters of spectra of Figure S5 according to equation S1<sup>a</sup>

Sample	Band	$\nu / \text{cm}^{-1}$	$\omega / \text{cm}^{-1}$	A	$I_0$	$I_{\text{max}}$	$I_D/I_G$	$I_G/I_{G'}$	$I_G/I_{G''}$
MWCNT	D	1346.9	49.9	66.8	0.156	1.01			
	G	1577.6	53.2	57.5	0.087	0.77	1.30	2.30	1.77
		1613.9	25.8	8.6	0.087	0.30			
	G'	2689.3	86.9	42.0	42.0	0.44			
MP50.2	D	1347.8	54.8	43.1	0.499	1.00			
	G	1579.2	52.0	43.2	0.464	0.99	1.01	1.37	1.37
		1614.9	25.8	6.4	0.464	0.62			
	G'	2691.1	87.3	29.8	0.509	0.73			
MP200.2	D	1347.1	51.9	48.1	0.408	1.00			
	G	1578.0	52.7	47.7	0.371	0.95	1.05	1.64	1.56
		1614.3	25.5	6.4	0.371	0.53			
	G'	2692.4	85.7	35.1	0.346	0.61			

<sup>a</sup>Fitting range in  $\text{cm}^{-1}$  for band D: 1270-1445, G: 1530-1650 and G': 2600-2800. The deviation of the fitted parameters were below 4.7%. Correlation coefficients were over 0.994.

#### Four-probes conductivity measurement

**Figure S6.** Voltage *versus* current for (a) MWCNT/P4VP; (b) MWCNT/metallopolymer composites.