Effect of Oxidation Approach on Carbon Nanotube Surface Functional Groups and Electrooxidative Filtration Performance

Supporting Information

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Table S1. Comparison of this study to previous studies on possible reaction sites of CNT.

Electrochemical Probes	Experiment methods	Anodic performance	Reaction site on CNT	Ref.
O_2, hydrazine, etc.	CV, TEM	Residual iron nanoparticles are responsible for the electrocatalytic oxidation properties	Nanoparticles	1-3
$Fe(CN)_6^{-3/-4}$ etc.	CV	Defects may play an important role in the electrochemisty of certain species, especially those that go via surface-mediated inner-sphere	Defects / oxidation states	4, 5
$Fe(CN)_6^{-3/-4}$, AA, etc.	CV	The relative electrosensitivity to the nanotube tip and sidewall and their oxidation states waries with different electrochemical probes	Tips / sidewall	6
$Fe(phen)_3^{2+/3+}$, $Ru(NH_3)_6^{3+}$ etc.	CV	Metallic and semiconducting CNT are sensitive to the redox couple	Conductivity	7
$Fe(CN)_6^{-3/-4}$, AA, phenol, oxalate.	CV, EIS, XPS, IR, TOC	This paper explored that effect of oxidation approach on carbon nanotube surface functional groups and how the specific CNT surface oxy-functional group affects the efficacy and efficiency of CNT anodes	Functional groups (-OH)	This paper

CNT Purification. Briefly, 1 g of as-received CNT was first calcinated in a tube furnace in the presence of O_2 by increasing from room temperature to 400 ^oC for at a rate of 5 ^oC per min and holding at 400 ^oC for 60 min (Thermolyne, 21100) to remove any amorphous or other non-CNT carbon impurities. Then, 0.5 g of calcinated CNT was placed into 0.5 L of concentrated HCl (37%) and heated to 70 ^oC in a round-bottom flask with a stir bar and a condenser for 12 hours to remove the residual metal catalyst impurities. After acid treatment, the sample was cooled to room temperature and vacuum filtered through a 5-μm PTFE membrane (Omnipore, Millipore) to collect the CNT. The CNT were then washed with MilliQ deionized water (DI) until the filter effluent pH was neutral.

Figure S1. Images of the Electrochemical Filtration Apparatus. The whole reaction cell (including the upper cap and bottom cap), anode components (Titamium ring and CNT network with cross-section and aerial images) and cathode (the perforated stainless steel).

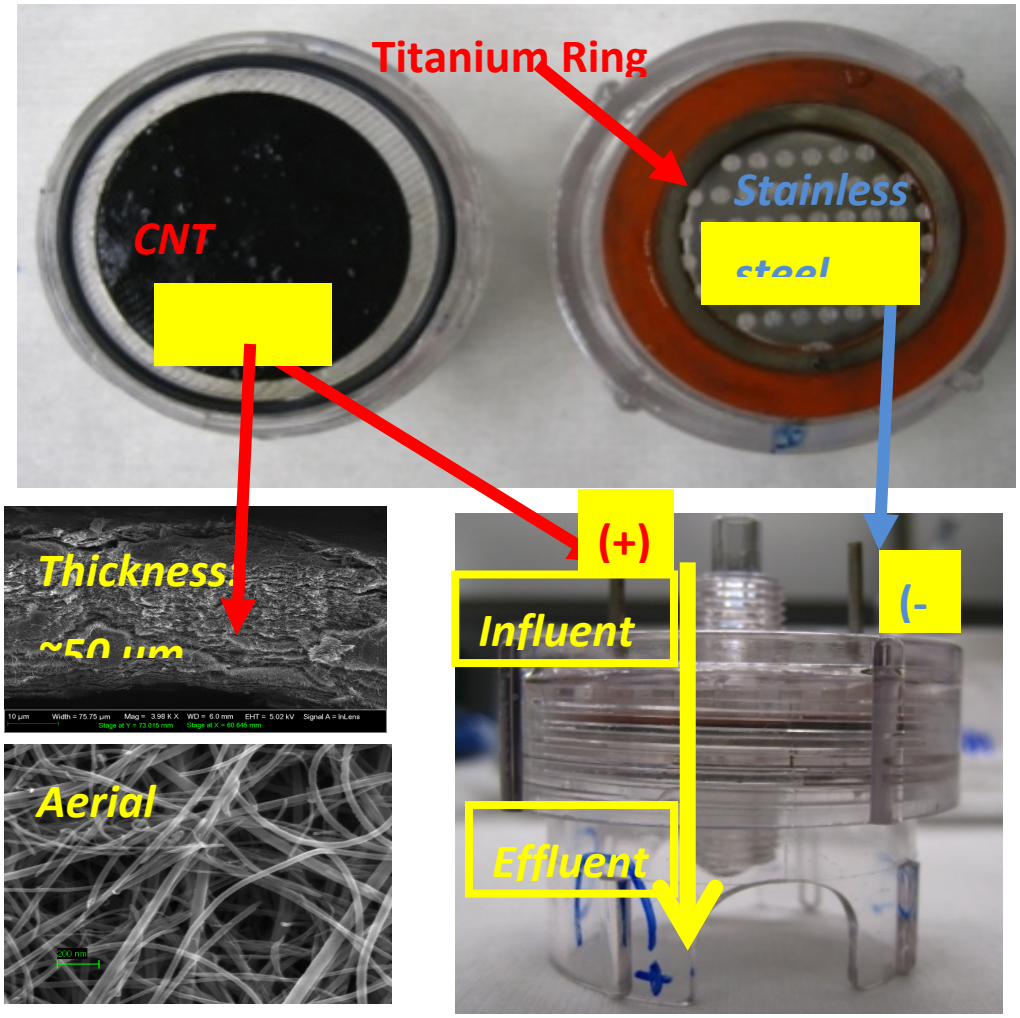
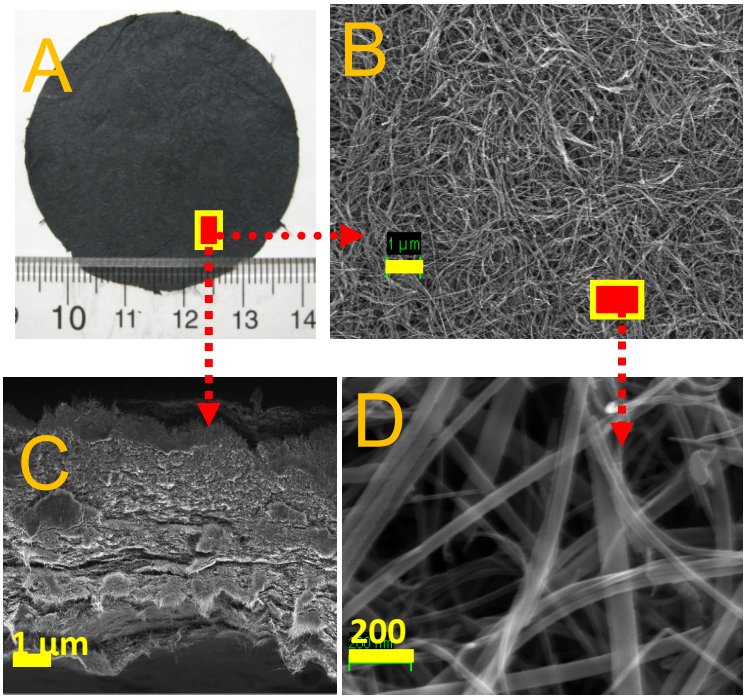


Figure S2. Images of a representative CNT network. A) Aerial image of CNT network. Scanning electron micrographs (SEM) **B)** aerial CNT network image; **C)** Cross-section CNT network image; **D)** magnified aerial CNT network image.



All information of XPS fitting (FWHM, Area under the curve etc.) is presented here.

XPS of **CNT-HCl**

Name	FWHM	Raw Area	%At Conc	c/C, o/O	O/C
C 1s	3.328	27.8428	6.701	0.0686	**0.023594079**
C 1s	1.125	247.937	59.465	0.6087	
C 1s	3.219	131.409	31.528	0.3227	
O 1s	4.39	5.23731	0.504	**0.2187**	
O 1s	2.606	18.7218	1.801	**0.7813**	

XPS of **CNT-EO**

Name	FWHM	Raw Area	%At Conc	c/C, o/O	O/C
C 1s	4.232	67.1179	7.678	0.079663	**0.037549**
C 1s	2.815	235.803	26.897	0.27907	
C 1s	1.105	542.082	61.806	0.641267	
O 1s	2.341	54.2417	2.482	**0.685825**	
O 1s	1.858	24.8869	1.137	**0.314175**	

XPS of **CNT-HNO_3**

Name	FWHM	Raw Area	%At Conc	c/C, o/O	O/C
C 1s	4.257	40.2416	9.94	0.103147	**0.03771**
C 1s	1.139	227.783	56.075	0.58189	
C 1s	3.097	123.245	30.352	0.314963	
O 1s	3.022	36.7945	3.634	**100**	

Table S1. BET Surface Area of CNT Samples.

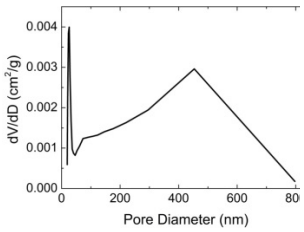
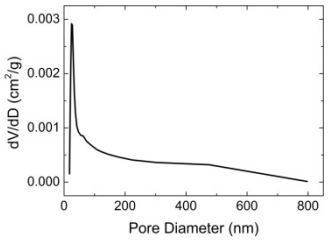
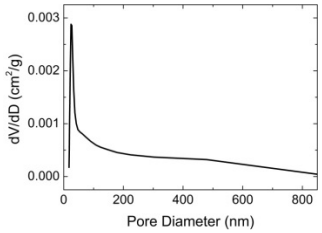
	CNT-HCl	CNT-EO	CNT-HNO_3
BET (m^2/g)	255.8	120.7	122.1
			

Figure S3. Time-dependent phenol degradation.

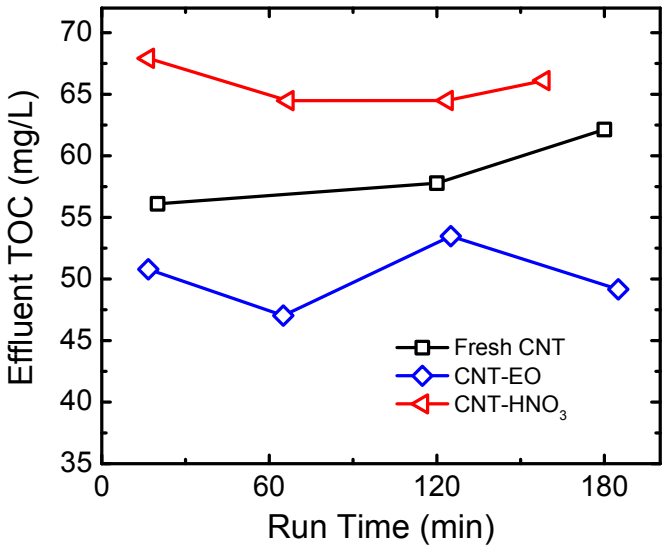
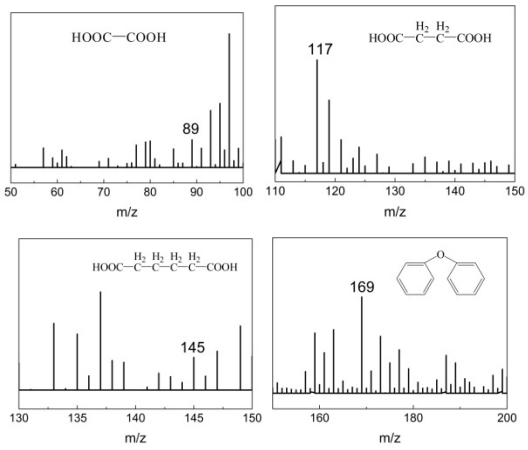


Figure S4. Intermediates of Phenol Electro-Oxidation Detected by UPLC-MS.



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