Supporting Information

Aging of Carbon Nanotubes Increases Their Adsorption towards Tetracycline

Xinxin Zhao [1,2], Huayu Liu [1,2], Zhen Yan [1,2] and Chao Song [1,2,*]

[1] Shandong Key Laboratory of Water Pollution Control and Resource Reuse, School of Environmental Science and Engineering, Shandong University, Qingdao 266237, China; zxx981260@163.com (X.Z.); liuhuayu0901@163.com (H.L.); yanzhen@email.sdu.edu.cn (Z.Y.)

[2] Shandong Key Laboratory of Environmental Processes and Health, School of Environmental Science and Engineering, Shandong University, Qingdao 266237, China

* Correspondence: songchao@sdu.edu.cn; Tel.: +86-532-58630936; Fax: +86-532-58630907

Dielectric Barrier Discharge Plasma Reactors

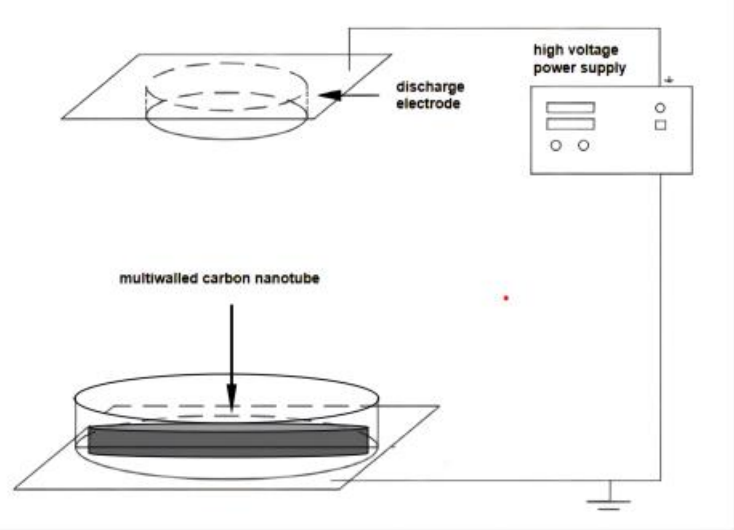

Figure S1. Schematic diagram of plasma treatment of CNTs.

The non-thermal plasma (NTP) device used in the experiment is shown in Figure S1, which consists of a discharge power supply and a plasma reaction vessel. The reaction vessel used in this study is a 10 mL culture dish, which is placed on a metal plate connected to ground electrode by a wire. The discharge high-voltage electrode is fixed on the reactor frame, and the bottom of the electrode is tightly connected with the insulating medium quartz glass sheet, which is placed directly above the culture dish. The distance between quartz glass sheet and culture dish is 0.5 cm.

Adsorption of tetracycline on CNTs

Adsorption kinetics experiment

Tetracycline solution (10, 20, 50 mg/L) were first prepared in DI water. Then, 0.15 g of pristine and aged CNTs were added in the solutions, respectively. The solutions were placed in a gas bath oscillator at 155 rpm at 28°C for 24h.

Adsorption isotherm experiment

TC solutions were prepared at the concentrations from 50 to 500mg/L. Then, 0.15 g of pristine and aged CNTs were added in the solutions, respectively. The solutions were placed in a gas bath oscillator at 155 rpm for 72h at 28°C and 48°C, respectively.

All experiments were repeated at least in triplicate, and the error bars represent the standard deviations calculated for each independent experiment.

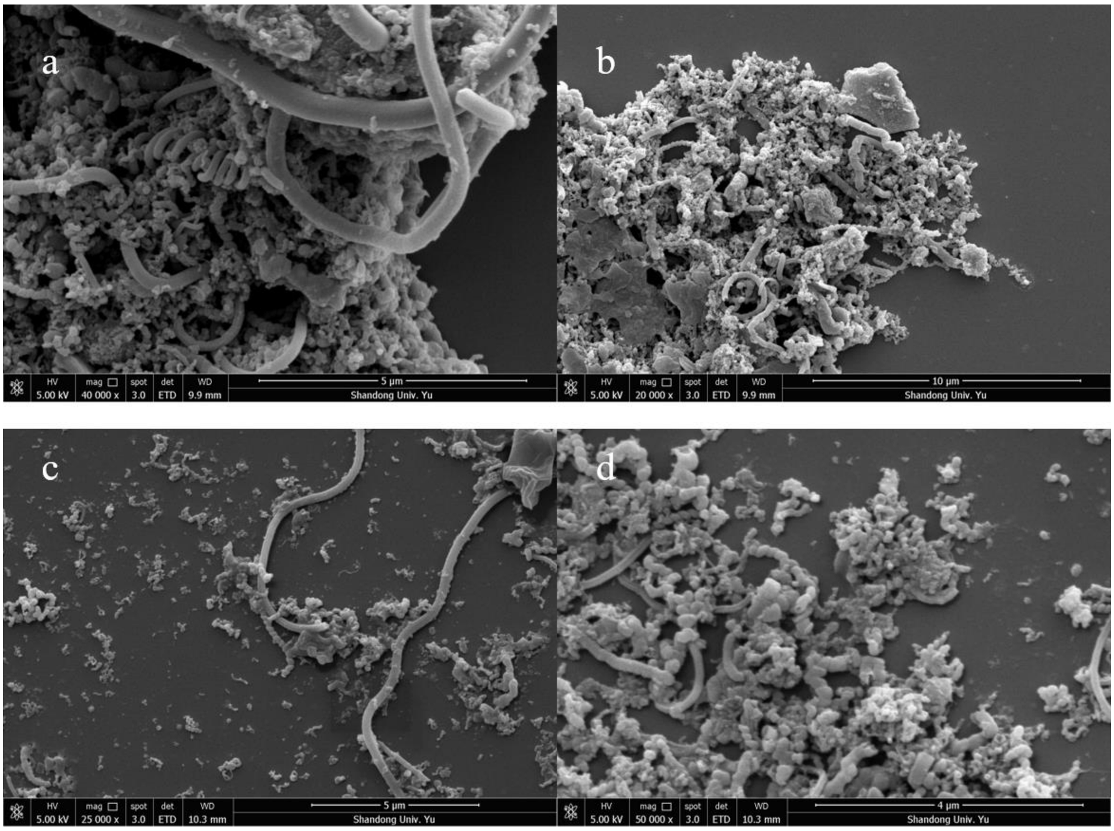

Figure S2. SEM characterization of pristine CNTs (a, b) and aged CNTs (c, d)

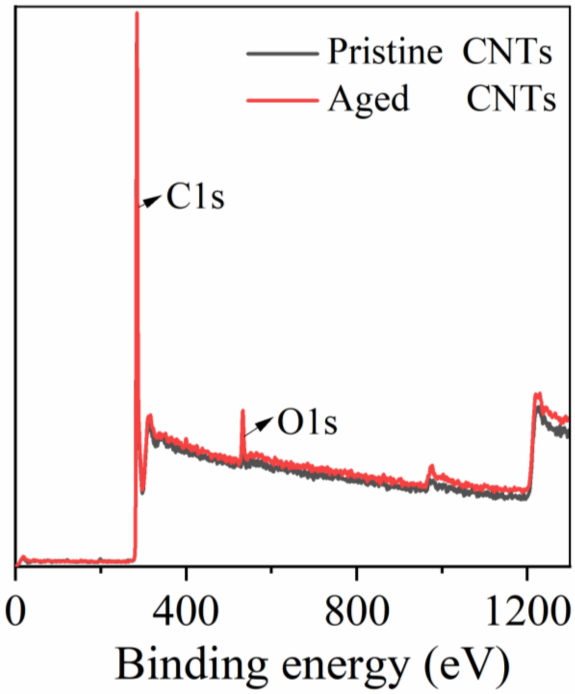

Figure S3. XPS spectra of pristine and aged CNTs

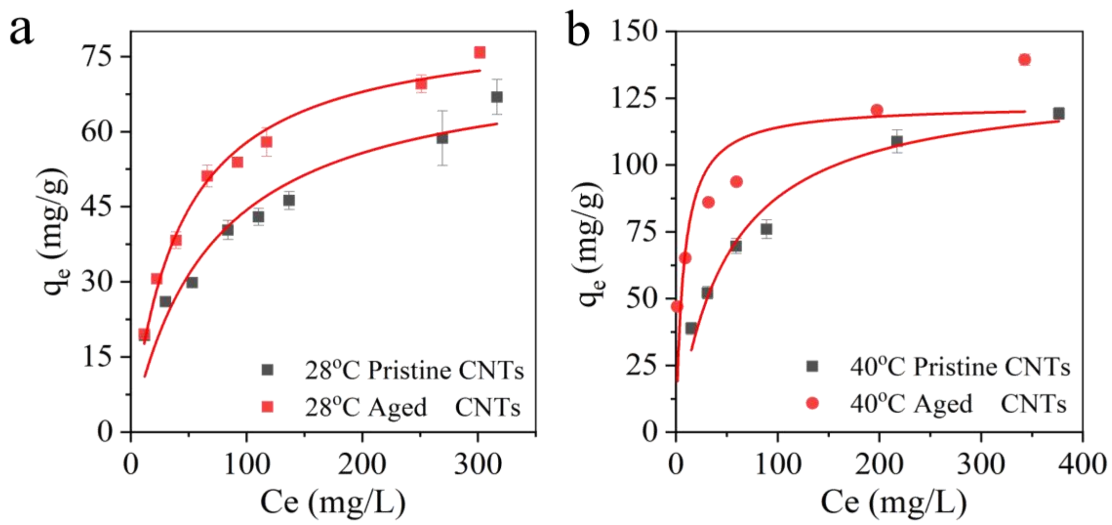

Figure S4. Adsorption isotherms fitted with Langmuir model of TC:(a)at 28℃; (b)at 40℃

Table S1. The chemical composition of the pristine and aged CNTs.

	C (atom %)	O (atom %)	C/O ratio
Pristine CNTs	98.12	1.88	52.19
Aged CNTs	96.18	3.82	25.18

Table S2. Kinetic parameters for the adsorption of tetracycline on the pristine and aged CNTs

Conc.[a] (mg/L)	Pseudo-first order kinetic				Pseudo-second order kinetic		
	$q_{e\ exp}$ (mg/g)	$q_{e\ cal}$ (mg/g)	k_1 (min^{-1})	R^2	$q_{e\ cal}$ (mg/g)	k_2 (g/mg/min)	R^2
Pristine CNTs −10	4.209	3.763	0.105	0.87	4.242	0.036	0.95
Aged CNTS −10	4.178	3.867	0.006	0.87	3.955	0.002	0.93
Pristine CNTs −20	7.860	6.639	0.080	0.79	8.325	0.014	0.89
Aged CNTs −20	7.743	7.351	0.005	0.95	7.049	0.0008	0.98
Pristine CNTs −50	18.932	15.745	0.036	0.74	18.631	0.003	0.85
Aged CNTs −50	17.510	15.636	0.005	0.93	16.748	0.0003	0.97

[a] Conc: the initial concentration of TC

Table S3. Isotherm parameters for tetracycline adsorbed by pristine and aged CNTs

	Langmuir Model			Freundlich Model		
	q_m (mg/g)	K_l (L/mg)	R^2	K_f ($mmol^{1-n}$ L^n kg^{-1})	n	R^2
Pristine CNTs −28ºC	75.012	0.014	0.911	6.836	2.559	0.987
Aged CNTs −28ºC	82.338	0.024	0.931	11.034	2.940	0.963
Pristine CNTs −40ºC	131.837	0.020	0.961	16.977	2.986	0.980
Aged CNTs −40ºC	122.604	0.133	0.662	42.141	4.955	0.995