Supplementary Materials

Carbon Nanotube-Based Nanocomposites in Environmental Remediation: An Overview of Typologies, Applications and an Analysis of Their Paradoxical Double-Sided Effects

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Tables

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Table S1. Main methods to synthesize carbon nanotubes (CNTs).

Method	Invention paternity/description	Type of tube	Advantages/Disadvantages	Refs
AD	(1991) From procedure to get fullerenes applying a current (110 A) at T >1,700°C CNTs form in the carbon soot of GR electrode	N.R.	⬇Structural defects Macroscopic production	[1–3]
LA	Dr. Richard Smalley* Graphite is blasted with a laser	MWCNTs	N.R.	[4,5] [6]
	Graphite and cobalt/nickel (metal catalyst particles) blasted with a laser	SWCNTs		[7]
CVD	Substrate of nickel, cobalt, iron, or combination catalyst NPs Heating at 700°C under a flux of a "process gas" ** and a "carbon-containing gas" *** to promote CNTs growing CNTs directly growth on the desired substrate by careful deposition of the catalyst	N.R.	⬇ Cost, scaling up Industrial production If present, need to remove the catalyst support via acid treatment, with possible damage to the CNT structure	[8–13] [8,14]
PECVD	Advanced CVD consisting of plasma generation by applying a strong electric field during CNTs growth	VACNTs	N.R.	[15]
HiPco	Rice University HPCO reacts with FePC, Fe NPs form providing the nucleation surface where CNTs form by the transformation of CO into carbon	SWCNTs	Production from milligrams to grams scale No environmental release of wastes	[4,5]
SGCVD	Kenji Hata, Sumio Iijima at AIST (Japan) Introduction of water into the CVD reactor	VANTAs Forest material	⬆Activity and lifetime of the catalyst ⬆To LA and HiPco by 100 times ⬇ Mass density, >99.98% pure, 2.5 mm height SWNT 10min Easy separation of CNTs from the catalyst >1 mm long VANTAs in several shapes §	[16] [5,17]
PT	Olivier Smiljanic (2000), Institut National de la Recherche Scientifique (INRS), Varennes (Canada) Argon, ethylene and ferrocene into PLASMOTRON, thus developing an intense 'flame' containing CNTs, metallic, carbon NPs and amorphous carbon	SWCNTs	10-fold ⬇consumed energy than in LA or AD.	[5,18]
ITP	Sherbrooke University and the National Research Council of Canada Modified PT procedure TP is generated by HFOCs in a loop and is conserved in flowing inert gas	SWCNTs	CNTs with different diameter distributions	[19]

LEM	Metal ions reduced to metal forms on cathode provide the nucleation surface for CNTs growth which derive from electrolysis of molten carbonates #	MWCNTs	⬆Valued CNTs Strategy for CO_2 capture and conversions	[5,20,21]
NICFE	CNTs form naturally in flames emitted by burning methane, ethylene, benzene		⬆Irregular in dimensions and quality CNT	[22–24]
ACVD	CNTs are synthesized in the gas phase and deposited in the form of randomly oriented networks, ready for many applications including transparent conductors.	SWCNTs TF	Clean defect less SWCNTs with limited yield	[25–27]

PR = Parameters; Adv = advantages; Disadv = disadvantages; * Rice University; AD = arc discharge; LA = laser ablation; CVD 0 chemical vapor deposition; ⬇ = less, minor, low; ** ammonia, nitrogen, or hydrogen; *** acetylene, ethylene, ethanol, or methane; PECVD = plasma-enhanced chemical vapor deposition; VACNTs = vertically aligned CNTs; HiPco = High-Pressure Carbon Monoxide Process; HPCO = high-pressure carbon monoxide; FePC = iron pentacarbonyl; SGCVD = super-growth CVD, also known as water-assisted chemical vapor deposition; VANTAs = millimetre-high vertically aligned nanotube arrays; "forests" materials = tubes aligned to the substrate; ACVD = aerosol CVD; TF = thin films; ⬆ = enhanced, improved, high, higher, highly, superior; § = sheets and bars, by applying weak compression during the process; PT = plasma torch; PLASMOTRON = microwave plasma torch; LEM = liquid electrolysis method; NICFE = natural, incidental, and controlled flame environments; GR = graphene; HFOCs = high-frequency oscillating currents; TP = thermal plasma; # the reactant is a carbon dioxide greenhouse gas.

Table S2. Most relevant research articles on CNT synthesis from biomass by different methods, operating parameters and main properties.

Feed stock	Method	Support/Catalyst	Operating parameters			CNTs	Properties				Refs.
			T (°C)	Time (min)	Carrier gas		SA (m^2/g)	D (nm)	*(%)	L (μm)	
Rice straw	P	Al_2O_3/Fe-Ni	830	30	N_2	MWCNT	188	15–40	N.A./N.A.	N.A.	[28]
Olive oil	P	Si wafer/$NiCl_2$	900	60	Ar	SWCNT	N.A.	27–31	N.A./ N.A.	N.A.	[29]
Turpentine oil	P	Zeolite/Fe-Co	850	25	N_2	SWCNT	N.A.	7–20	58/N.A.	N.A.	[30]
Coconut oil	CVD	Fe	850	60	N_2	MWCNT	N.A.	80–100	58/N.A.	3–4	[31]
Sesame oil	P+CVD	FNS/CH_3CN	900	15	Ar	MWCNT	N.A.	30–60	58/N.A.	3–4	[32]
Palm oil	CVD	Silicon/Ferrocene	750	30	Ar	SWCNT MWCNT	N.A.	0.6–1.2	90/N.A.	110	[33]

Plastic waste	P+CVD	Ni	800	30	N_2	MWCNT	N.A.	40–50	N.A./31	N.A.	[34]
Petroleum coke	CVD	Silica/Fe	700	60	He	MWCNT	N.A.	18	N.A./N.A.	N.A.	[35]
Plastic waste	P	Cordierite/*N*-Mg	750	60	N_2	MWCNT	N.A.	30–50	N.A./93	30–50	[36]
NCG	P+CVD	Ni/Al_2O_3	1000	112	N_2	SWCNT	N.A.	10	N.A./82	N.A.	[37]
Ethylene	FC, CVD	Co/Fe, Co/Ni, Ni, Co, Fe	1050	0.17	N_2	SWCNT	N.A.	0.67–2	61–69/ N.A.	N.A.	[38]
Rice straw	T-CVD	Fe, Ni	800	120	N_2	MWCNT	20 (CNT+Fe)35 (CNT+Fe+Ni)	22 (CNT+Fe)66 (CNT+Fe+Ni)	N.A./41–44	N.A.	[39]

*Purity/yield; CH_3CN = acetonitrile; N.A. = information not available; D = Diameter; P = pyrolysis; SA = surface area; T = temperature; NCG = natural condensed gas; floating catalyst; CVD = chemical vapor deposition; T-CVD = thermal CVD; FNS = ferrocene nitrogen source.

Table S3. Preparation of CNTs from biomass via microwave irradiation.

Raw material	Microwave conditions	Product	Properties of CNTs	Refs
Sugarcane bagasse	600 W, 500 °C	NPs and CNTs	Average Ø = 20-50 nm	[40]
Waste rice husk	900 W, 2.45 GHz	GR CNTs + GR-CNTs	L = tens of μm, Ø = 50-200 nm	[41]
Wheat straw, oat husk, rapeseed cake and hazelnut hulls	200 W, 2.45 GHz, 80 °C, 17 psi	CNTs	Ø = 17-100 nm	[42]
Palm kernel shells	2000 W, 2.45 GHz, 600 °C	MWCNTs	Ø = 50-100 nm	[43]
Pine nut shells	2000 W, 600 °C	MWCNTs	L = 2600-3200 nm	[44,45]
Gumwood	300 W, 500 °C, 2.45 GHz	MWCNTs	WT = 5-7 nm, Ø about 50 nm	

L = length; Ø = diameter; NPs = nanoparticles; Gr = graphene; WT = wall thickness.

Table S4. Reported applications of CNTs and modified CNTs for absorption of several heavy metals from water by different mechanisms.

Target pollutant	Adsorbents	Adsorption Mechanism	pH, CT, AD*	Refs.
Hg (II)	ATPPBr-CNT	IE/complexation	pH 5.5, 28 min, 5.5 mg	[46]

	TBABr-Gly-CNT	Physisorption	pH 6.4, 45 min, 6.0 mg	[47]
	MWCNTs-AA	Chemisorption	pH 6.0, 180 min, 10 mg	[48]
	CNTs- MnO_2	EI	pH 5–7, 80 min, 20 mg	[49]
	SWCNT-Thiol (SH)	Chemisorption	pH 5.0, 60 min, 250 mg	[50]
	MWCNTs-Sulphur	SASBI, EI	pH 12.15, 60 min, 100 mg	[51]
	O-MWCNTs	Chemisorption, ED, IE, physisorption, complexation	pH 6.0, 90 min, 20 mg	[52]
	CNTs-Iodide	Chemisorption, ED, IE, physisorption, complexation, SASBI	pH 6.0, 90 min, 20 mg	[52]
	MWCNTs-Sulphur	IE, physisorption, chemisorption, ED, complexation, SASBI	pH 6.0, 90 min, 20 mg	[52]
	Amino, SH-MWCNTs	Physisorption, SASBI	pH 6.0, 60 min, 400 mg	[53]
	Raw MWCNTs	EI, complexation, chemisorption, IPD	pH 3.5, 10 mg	[54]
	MWCNTs-C_6H_5OH	ED	pH 1.2, 10 mg	[54]
	MWCNTs-COOH	ED	pH < 1.0, 10 mg	[54]
	MWCNTs	Chemisorption	pH 7.0, 500 mg, 120 min	[55]
	PAAA/GO-SWCNTs	Chemisorption	pH 5.3, 210 mg, 6.87 min	[56]
	MWCNTs-COOH	Chemisorption, EI	pH 7.0, 500 mg, 90 min	[57]
	MWCNTs-OH	Chemisorption, EI	pH 7.0, 500 mg, 90 min	[57]
	MWCNTs-NH_2	Chemisorption, EI	pH 7.0, 500 mg, 90 min	[57]
	MWCNTs-3-AP	Chemisorption, physisorption	pH 7.7, 20 mg, 120 min	[58]
	CNTs-SH/Fe_3O_4	Chemisorption, physisorption	pH 6.0, 500 mg, 90 min	[59]
	CuS-MWCNTs	Chemisorption, physisorption	50 mg,120 min	[60]
	MWCNTs-KOH@NiNPs	Chemisorption	pH 5.5, 40 mg, 30 min	[61]
Pb (II)	CNTs-DES	Chemisorption, EI	pH 5, 5 mg, 15 min	[62]
	T2AEA-MWCNTs	Chemisorption, physisorption	pH 6.6, 10 mg, 45 min	[63]

	M-MWCNTs- 8-aminoquinline	Chemisorption	pH 6.4, 25 mg, 5 min	[64]
	SWCNTs-WSh	Chemisorption, precipitation	pH 5, 1000 mg, 30 min	[65]
	As-produced MWCNT	Chemisorption, EI	pH 5.5, 20 mg 120 min	[66]
	O-MWCNTs	Chemisorption, EI	pH 5.5, 1000 mg 120 min	[66]
	O-MWCNTs	EI	pH 5, 5 mg, 30 min	[67]
	MWCNTs-isocyanate	EI	pH 6, 5 mg, 30 min	[68]
	PP-MWCNTs	Chemisorption, physisorption	pH 6, 60 mg, 60 min	[69]
	N_2H_4-SH-Fe_3O_4/O-MWCNTs	Chemisorption, physisorption	pH 6, 40 mg, 30 min	[70]
	Fe_3O_4/O-MWCNTs	Chemisorption, complexation, EI	pH 6, 1000 mg, 360 min	[71]
	PAMAM-CNTs	Physisorption, EI	pH 7, 30 mg, -	[72]
	Acidified MWCNTs	Chemisorption, complexation, IE	pH 9, 500 mg, 600 min	[73]
	Fe_3O_4-CNT	Chemisorption, physisorption	pH 6, 100 mg, 40 min	[74]
	NiO_2–MWCNTs	Chemisorption, physisorption	pH 7, 2000 mg, 10 min	[75]
	DIC-O-MWCNTs	Chemisorption	pH 5, 250 mg, 120 min	[76]
	O-MWCNTs	Chemisorption	pH 6, 200 mg, 80 min	[77]
	MWCNTs@SiO_2-NH_2	Chemisorption	pH 5, 20 mg, 60 min	[78]
	MWCNT-SH	Chemisorption	pH 5, 100 mg, 5–40 min	[79]
	SPP-MWCNTs	Chemisorption	pH 5, 10 mg, 60 min	[80]
	O-MWCNTs	Chemisorption, EI	pH 5, 30 mg, 180 & 360 min	[81]
	O-MWCNTs	Chemisorption	pH 5, 10 mg, 10 min	[82]
	CNT–steel slag	Chemisorption, IE, precipitation	pH 6.5, 20 mg, 90 min	[83]
	CoBi-LDH-Cr@CNT	Chemisorption	pH 7, 5 mg, 180 min	[84]
	MWCNT-Au/Fe_3O_4	Van der Waals, EI, IE, complexation	pH 7, 100 mg, 100 min	[85]
As (V)	O-MWCNTs	Chemisorption, EI	pH 4, 100 mg, 45 min	[86]

	Ce-Fe-MWCNTs	Chemisorption, EI	pH 4, 10 mg, 360 min	[87]
	Fe $(NO_3)_3$-CNT	Chemisorption, physisorption, Coprecipitation	pH 5-6, 100 mg, 120 min	[88]
	ZVI-MWCNTs	Chemisorption, complexation	pH 7, 2500 mg, 240 min	[89]
	Fe_2O_3-SWCNTs	Chemisorption	pH 4, 100 mg, 3 min	[90]
	CF-MWCNTs	Chemisorption	pH 6.5, 2000 mg, 240 min	[91]
	Zn-BDC@CT-CNT	Chemisorption	pH 4.0,10 mg, 20 min	[92]
	MWCNTs-KOH@NiNPs	EI, surface adsorption, IE, pore diffusion	pH 5, 40 mg, 30 min	[61]
	Co-Fe-N-CNTs	Coprecipitation	pH 4.5100 mg, 30 min	[93]
	Ce-Fe-MWCNTs	Chemisorption, complexation	pH 7.5, 10 mg, 360 min	[87]
	ZVI-MWCNTs	Chemisorption, complexation	pH 7, 2500 mg, 240 min	[89]
	CNTs-DES	Chemisorption	pH 2.7, 10 mg, 30 min	[62]
As (III)	MA-MWCNTs	Chemisorption, liquid film diffusion	pH 6.0, 2000 mg, 60 min	[94]
	$ZrO(OH)_2$-CNTs	Chemisorption, EI	pH 7, 150 mg, 360 min	[95]
	MWCNTs-3-AP	Chemisorption, physisorption	pH 7.7, 20 mg, 120 min	[96]
	CNTs–PAMAM–Ag	Chemisorption, IPD, boundary layer effect	pH 8100 mg, 15 min	[97]
	S-MWCNTs	Physisorption, EI	pH 6, 25 mg, 360 min	[98]
	SWCNTs-OH-RGO	π–π interactions	pH 6.8, < 50 mg, 180 min	[99]
	DTCA-MWCNTs	Chemisorption, physisorption	pH 6, 5.0 mg, 120 min	[100]
	PHB-CNTs	IE, EI	pH 5.65, 20 mg,10 min	[101]
Cu (II)	NN-mSiO_2-MWCNTs	Chemisorption	pH 6.2, 20 mg, 30 min	[102]
	Ag-MWCNTs	Chemisorption, IE	pH 6, 50 mg, 100 min	[103]
	CS-MWCNTs	Chemisorption, physisorption	pH 5.5, 50 mg, 90 min	[104]
	As-produced MWCNT	Chemisorption, EI	pH 5.5, 20 mg 120 min	[103]
	O-MWCNTs	Chemisorption, EI	pH 5.5, 1000 mg 120 min	[66]

	O-MWCNTs	Chemisorption, EI	pH 6, 30 mg, 75 min	[105]
	O-MWCNTs	EI	pH 5, 5 mg, 30 min	[106]
	MWCNT-TA	Charge interaction	pH 6, 10 mg, 240 min	[107]
	ACh-CNTs	Chemisorption, surface diffusion	pH 7, 50 mg, 300 min	[108]
	Acidified-MWCNTs	Chemisorption, IE, complexation	pH 9, 50 mg, 600 min	[73]
	M-MWCNTs	Chemisorption, surface diffusion, EI	pH 7, 100 mg, 30 min	[109,110]
	TR-CNTs	EI, pore filling, H-bonding, complexation.	pH 6, 400 mg, 60 min	[111]
	PAAm/FMWCNTs	Chemisorption, complexation	pH 5, 1000 mg 90 min	[112]
	THF-CNTs	Chemisorption	pH 5, 200 mg 30 min	[113]
	DA-MALI-CNTs	Chemisorption, chelation	pH 7, 10 mg, 60 min	[114]
	O-MWCNTs	EI, π–π interaction	pH 6, 240 min	[115]
	Double-O-MWCNTs	Chemisorption	pH 7, 20 mg, 840 min	[116]
	CNF/MWCNTs/SnO_2	Chemisorption, EI	pH 6,50 mg, 30 min	[117]
	MWCNT-PEI	Chemisorption	pH 7,10 mg, 180 min	[118]
	MWCNTs-PEG-PVA	Chemisorption, physisorption	pH 6, 20 mg, 80 min	[119]
	MWCNT-RAFT	Complexation, EI	pH 7, 1 mg, 180 min	[120]
Cd (II)	Al_2O_3-MWCNTs	EI, physisorption, surface precipitation, surface complexation	pH 7, 50 mg, 240 min	[121]
	Acid modified CNTs	EI	pH 7, 50 mg, 240 min	[122]
	Ag-MWCNTs	Chemisorption, IE	pH 7, 50 mg, 100 min	[103]
	O-MWCNT	Chemisorption, EI	pH 5.5, 1000 mg 120 min	[66]
Empty Cell	Raw CNTs	EI	pH 4, 75 mg, 240 min	[123]
	PP-MWCNTs	Chemisorption, physisorption	pH 6, 60 mg, 60 min	[69]
	MWCNTs-COOH	Chemisorption, EI	pH 6, 60 mg, 60 min	[69]

	Acidified- MWCNTs	Chemisorption, IE, complexation	pH 9, 50 mg, 600 min	[73]
	M-MWCNTs-8-AQ	Chemisorption	pH 6.4, 25 mg, 5 min	[64]
	O-CNTs	Chemisorption, surface diffusion	pH 4, 40 mg, 300 min	[124]
	Pure CNTs	Chemisorption, physisorption	pH 10,500 mg, 500 min	[125]
	NiONPs-MWCNTs	Chemisorption, EI	pH 7-8, 200 mg, 20 min	[126]
	Modified MWCNTs	Chemisorption, physisorption	pH 5, 1000 mg, 1440 min	[127,128]
	MWCNTs-KOH@NiNPs	EI, surface adsorption, IE, pore diffusion	pH 5, 40 mg, 30 min	[61]
	PA-CNT	EI, complexation, π–metal interactions, FGI	pH 7, 30 mg, -	[129]
	MWCNTs	Chemisorption, physisorption, EI	pH 4, 100 mg, 720 min	[130]
	NCs	Chemisorption, physisorption, EI	pH 2, 2500 mg, 240 min	[131]
	O-MWCNT	Chemisorption, EI	pH 5.5, 1000 mg 120 min	[66]
	IL-oxi-MWCNTs	EI, cation/anion-interaction	pH 2.5-4.0, 150 mg, 40 min	[132]
	Raw CNTs	EI	pH 4, 75 mg, 240 min	[133]
	pTSA-Pani@GO-CNT	EI, π–π interaction	pH 2, 200 mg, 500 min	[134]
	M-MWCNTs	Chemisorption	pH 3, 100 mg, 700 min	[135]
	SM-MWCNTs	IPD	pH 1, 5 mg, 60 min	[136]
Cr (VI)	EDTA/H_2SO_4- MWCNTs	Chemisorption, IPD	pH 3, 60 mg, 150 min	[137]
	CC-CNTs	Chemisorption, physisorption	pH 3, 10 mg, 660 min	[138]
	Fe_3C-CNTs	EI, complexation	pH 5.4,10 mg, 1440 min	[139]
	PA-CNTs	Redox, complexation, EI	pH 4, 400 mg, 3 min	[139]
	CaO_2-CNTs	Chemisorption	pH 3, 200 mg, 120 min	[140]
	MWCNTs-Ag0 /PVAc	Chemisorption	pH 3, 40 mg, 60 min	[61]
	AOMW-MWCNTs	EI, chemisorption, IE	pH 2, 25 mg, 120 min	[141]
	mZVI-CNTs	EI, H-bonding, reduction, precipitation	pH 5,1000 mg, 2100 min	[142]

Zn (II)	Ch/MWCNTs-COOH	Reduction, EI	pH 2, 50 mg, 30 min	[143]
	Ni@N-CNT	Reduction, EI	100 mg, 15 min	[144]
	CNTs	Chemisorption, EI	pH 10, 900 mg, 120 min	[145]
	MWCNTs	EI	pH 10, 90 mg 120 min	[146]
	O-MWCNTs	EI	pH 5, 5 mg, 30 min	[106]
	Fe_3O_4/O-MWCNTs	Chemisorption, complexation, EI	pH 6, 1000 mg, 360 min	[70]
	MWCNTs	Chemisorption	pH 5,60 mg, 360 min	[63]
	MWCNTs	Chemisorption, IE, EI	pH 10, 50 mg, 60	[145]
	f-MWCNT	Surface complexation	pH 7, 20 mg, 60 min	[147]
	Ar/O_2-CNTs	Surface complexation	pH 7, 29400 mg, 60 min	[148]
	O-CNTs	Chemisorption, EI	pH 6, 50 mg, 100 min	[63]
Ni (II)	As-produced MWCNT	Chemisorption, EI	pH 5.5, 20 mg, 120 min	[66]
	O-MWCNTs	Chemisorption, EI	pH 5.5, 1000 mg, 120 min	[66]
	PP-MWCNTs	Chemisorption, physisorption	pH 6, 60 mg, 60 min	[69]
	PAMAM/CNTs	Chemisorption, physisorption, EI, chelating	pH 7, 30 mg, 15 min	[149]
	Acidified MWCNTS	Chemisorption, complexation, IE	pH 9, 500 mg, 600 min	[73]
Co (II)	MWCNTs	Chemisorption, IPD, EI	pH 8, 5000 mg, 300 min	[150]
	MWCNTs	Chemisorption	pH 5, 30 mg, 60 min	[151]
	MWCNTs-HAP	Surface complexation	pH 6, 60 mg, 20 min	[151]
	PAMAM/CNTs	Chemisorption, physisorption, EI, chelating	pH 7, 30 mg, 15 min	[148]
Empty Cell	NaAlg-CNTs	EI	pH 6.8, 100 mg, 540 min	[152]
	SMMWCNTs	EI, complexation	pH 6, 4.5 min, 300 mg	[153]
	MWNCTs	EI	pH 10, 38.6 min, 1.57 mg	[154]

* Optimized conditions; CT = contact time; AD = absorbent dose expressed in mg/L; ATPPBr = allyl triphenyl phosphonium bromide; TBABr = tetra n-butyl ammonium bromide; PAAA = poly-(allyl acetoacetate); GO = graphene oxide; ChC = chitosan-coated; 3-AP = 3-aminopyrazole; SM = super

magnetic; Gly = glycerol; AA = amido amine; PVAc = polyvinyl acetate; DES = deep eutectic solvent; T2AEA = tris-(2-aminoethyl)-amine; M-MWCNT = magnetic MWCNT; 8-AQ = 8-aminoquiline; PP = poly pyrrole; O-MWCNT = oxidised MWCNT; PAMAM = poly amido amine; DIC = di-iodo-carbene; SPP = seleno phosphoryl; ZVI = zero valent iron; CF = carbon foam; MA = microwave-assisted; RGO reduced graphene oxide; DTCA = di thiocarbamate; PHB = poly-hydroxyl butyrate; Ach = acid chitosan; IE – ionic exchange; EI = electrostatic interactions; ED = external diffusion; SASBI = soft acid soft base interactions; IPD = intraparticle diffusion; FGI = functional groups interactions.

Table S5. CNTs as adsorbent materials to remove heavy metals in wastewater treatment.

Adsorbent	Synthesis Method	Xenobiotic	Operating parameters	Removal efficiency/Adsorption capacity	Refs.
MI MWCNT	N.A.	Cu (II)	CT = 35 min, DA = 100 mg, pH = 5.5, AS = 160 r/min IM= Freundlich and Langmuir, KM = PSO	99.9%	[155]
MWCNT	TMW CVD	Pb (II)	pH = 5, IC Pb (II) = 10 mg/L, AS = 2.8 r/*sec* DA = 100 mg, CT = 22.5 min IM = Langmuir and Freundlich, KM = PSO	99.9%	[156]
F-MWCNT	N.A.	Cu (II)	CT = 1 h, DA = 10 mg/L, IC = 20 mg, pH = 3 IM = Langmuir, KM = PSO	93%	[157]
CNT/WCPO/Fe	Thermal CVD	Zn (II), Cu (II) Fe (II)	DA = 1800 mg/L, pH = 7, IC heavy metal = 100 mg/L IM = Langmuir for Cu (II)	99.2-99.9%	[158]
MoS_2+TF-MWCNT	TMW CVD	Pb (II), Cd (II)	IM = Freundlich, KM = PSO, DA = 2 mg/mL, pH = 6 CT = 1 h	Cd (II) = 66.6 mg/g, Pb (II) = 90 mg/g	[159]
MWCNT-COOH MWCNT-OH	Catalytic CVD	Cr (VI)	IC Cr (VI) = 5–60 mg/L, pH = 5, DA = 25 mg KM = IPDM and PSO	8.09 mg/g (MWCNT-COOH) 7.85 mg/g (MWCNT-OH)	[160]

N.A. = Not available; AD = adsorbent dose; F = functionalized; TF = thiol-functionalized; CVD = chemical vapor deposition; TMW CVD = tubular microwave CVD; AS = agitation speed; IM = isotherm model; KM = kinetic model; IC = initial concentration; CT = contact time; PSO = pseudo second order kinetic; IPDM = intraparticle diffusion kinetic model.

Table S6. Applications of CNTs for the removal of pesticides, pharmaceuticals and some other emerging organic pollutants by adsorption.

Target xenobiotic	Adsorbent	MAC (mg/g) or RE (%)	Optimized condition	Ref.
1PBA, DDB	SWCNTs	50% (1PBA), 28% (DDB)	↑CLD, TH = 50 μm; Xenobiotic = 15 μg/mL	[161]
Oxytetracycline (OXY)	MWCNTs	450 mg/g	pH 7, 1560 min,100 mg	[162,163]
Ibrupofen	MWCNTs	9 mg/g	pH 4, 2.4 mg	[164]
	MWCNTs	12 mg/g	pH 4, 30 min, 50 mg	[165]
Tetracycline	MWCNTs	42 mg/g	pH 10, 2.4 mg	[164]
Empty Cell	CNTs–C@Fe–CS	104 mg/g	pH 10, 1440 min, 100 mg	[106]
Empty Cell	Fe_2O_3@OCNT	96%	pH 6, 0.13 min	[166]
Sulfamethazine	MWCNTs	34 mg/g	pH 5, 1440, 320 mg	[167]
Basic Red 46	SWCNT-COOH	49 mg/g	pH 9,10 min, 50 mg	[168]
Carbamazepine	GCNTs	87 mg/g	pH 7, 4320 min,150 mg	[169]
Diclofenac sodium	GCNTs	65 mg/g	pH 4, 4320 min,150 mg	[169]
Fulvic acids	H_2O_2-MWCNT	322 mg/g	30 min, 2 mg	[170]
Atrazine	MCNTs	58 mg/g	pH 7, 180 min, 500 mg	[171]
Azorubine	Fe_2O_3-MWCNT	> 95%	pH 5, 20 min, 130 mg	[172]
Diquat dibromide	MWCNTs	58 mg/g	pH 6.5, 300 min, 10 mg	[173]
Toluene	(MWCNTs)-SiO_2	50 mg/g	pH (2-3), 30 min, 1000 mg	[174]
Cymoxanil	CNT-COOH/MnO_2/Fe_3O_4	11 mg/g	pH 10, 5 min, 5 mg	[175]
Pirimicarb	CNT/Fe_3O_4	95%	pH 6.72, 47.56 min, 1070 mg	[176]
Malathion	F-MWCNTs	98%	pH 12, 50 min, 5 mg	[177]

1-PBA = 1-pyrene butyric acid; DDB = diquat dibromide; MAC = Maximum adsorption capacity; RE% = removal efficiency percentage; F-MWCNT = functionalized MWCNT; GCNT = granular carbon nanotube; MCNT = magnetic carbon nanotube; optimized conditions refer in the order to values of pH, contact time and adsorbent amount; CLD = cross-linked degree; TH = thickness; ↑ high, higher; @ = activated.

Table S7. CNTs as adsorbent materials to remove dyes from wastewater (WW).

Adsorbent	Synthesis Method	Xenobiotic	Operating parameters	Removal efficiency/Adsorption capacity	Refs.
SWCNT	ADSC/Crosslinking	MB	↑Cross-linking degree, thickness = 50 μm Xenobiotic = 15 μg/mL	83% (MB)	[161]
MWCNT	Commercial	CR, PR dyes	pH = 6, T = 25° C, dye = 200 mg/L, dose = 1 g/L	PR = 68 mg/g, CR = 256 mg/g	[163]
MWCNT	Commercial	CR	CT = 1 h, pH = 11, T = Endothermic IC = 200 ppm, MWCNT dose = 50 mg	92%	[178]
MWCNT	Catalytic CVD	Yellow 81, Red 159 Blue 116	IC dye = 100 mg/L, CNT = 60 mg, PSO IM: Blue 16 = Temkin IM: Red 159 = Langmuir/Freundlich IM: Yellow 81 = Freundlich/Temkin	33 mg/g	[179]
MWCNT	Catalytic CVD	MB, MO	CT = 1 h, catalyst dose = 20 mg, IC dye = 10 mg/L T = 298 K, pH = 6, KM = PSO	MB = 7 mg/g, MO = 5.5 mg/g	[180]
Chitosjan+SiO_2+MWCNT	Gelation method	RB19, DB 71	pH = 6.8 (DB71), pH = 2 (RB 19), IM = Langmuir KM = PSO, R^2 = 0.996 (DB71), 0.998 (RB19)	DB71 = 61.4 mg/g, RB19 = 97.1 mg/g	[181]
CNTs.	N.A.	DR 1, AB 113 MB, MO	KM = PSO, IM= Langmuir, IC dye = 20 mg/L, T= 25°C Dose (mg/g) = 19.4 (DR1 and MB 18.4 (AB113), 19 for (MO) pH = 8 (MB), 7 (AB113), 6 (MO), 7 (DR1) CT = 15 min (MB, MO, AB113), 10 min (DR1)	DR1 = 500 mg/g, MB = 91 mg/g MO = 96 mg/g, AB113 = 172 mg/g	[182]
Fe_3O_4/AA/IPAA/MWCNTs	N.A.	MB, RhB, CV	KM = PSO, IM = Langmuir, IC dye = 50 mg/L AD = 50 mg, pH = 8, T = 25°C	CV = 287 mg/g, MB = 302 mg/g RhB = 231 mg/g	[183]

Gel/F_2O_3/MWCNT	Co-P/EM	MB, DR 80	IM = Freundlich, KM = PSO, T = 65°C, time = 6 h IC = 500 mg/g	DR81 = 96%, MB = 76%	[184]

N.A. = Not available; gel = Gelatine; AA = acrylic acid; IPAA = isopropyl acryl amide; MI = microwave induced; AD = adsorbent dose; ADSC = aryl diazonium salt chemistry; F = functionalized; TF = thiol-functionalized; CVD = chemical vapor deposition; TMW CVD = tubular microwave CVD; Co-P/EM = co-precipitation/emulsification; AS = agitation speed; IM = isotherm model; IC = initial concentration; CT = contact time; T = temperature; ↑ high, higher; PSO = pseudo second order kinetic; IPDM = intraparticle diffusion kinetic model; PR = ponceau 4R dyes; RB19 = reactive Blue 19; DR71 = direct Blue 71; DR1 = disperse red 1; AB113 = acid blue 113; CV = crystal violet; DR80 = direct red 80; WCPO = waste cooking palm oil; KM = kinetic model.

Table S8. Application of CNTs for the photocatalytic oxidative degradation of organic contaminants in WW.

Photocatalyst	Methods	Light source	Xenobiotic	Operating parameters	Removal efficiency	Refs.
MWCNT/TiO_2	Sol-gel	Vis	MB	MWCNT/TiO_2 molar ratio (wt) 0.05/1 LI = 15 W, CT = 400 C, time = 180 min, XC = 1×10^{-5} M	TiO_2 only 22% MWCNT+TiO_2 70%	[185]
MWCNT/C_3N_4	Hydrothermal	Vis	RhB, MB and MO	LI = 300 W, $\Lambda \geq 400$ nm, CD = 5 mg/mL IT = 1.5 h (MB), 3 h (MO and RhB)	MB 67%, MO 90%, RhB 85%	[186]
CNT/P-TiO_2	Hydrothermal	UV and Vis	MO	Mass ratio CNT/P-TiO_2 5/100, LI UV = 250 W LI Vis = 400 W, IT = 80 min	~100%	[187]
CNT/g-C_3N_4/$BiVO_4$	Wet impregnation	Simulated sunlight	Phenolic compounds	IR = 2 h, PFO, Temkin model, PC = 10 mg/L, LI = 500 W	81%	[188]
CNT/TiO_2/Al_2O_3	Hydrothermal	UV	Metamifop herbicide	CD = 10 mg, XC = 5 ppm	85%	[189]
CNT/TiO_2/PAN	Electro-spinning	UV	Phenol	pH = 5, IT = 7 min, LI = 100 W, CD = 20 mg PC = 10 ppm, R&R = 3 cycles	99%	[190]
CuO/CNTs	Chemical method	UV	DR and RR	LI = 9 W, FO, RR 0.0105 (RR120), 0.0137 (DR31) min^{-1} CD = 5 mg	RR 87%, DR 89%	[191]
Ti^{3+}/TiO_2/SWCNT	CP + calcination	Vis	MB	IT = 25 min, RR = 0.0083 min^{-1}	83%	[192]

MWCNT/TiO_2/SiO_2	Sol-gel	UV	CBZ and BPA	PFO RC = 0.0131–0.0743 (CBZ), 0.0827–0.1751 (PBA) min^{-1} IT = 20 min, CC = 500 mg/L	50%	[193]
MWCNT/Ag-ZnO	Precipitation	Vis	CR	CC = 150 mg/L, LI = 40 W, pH = 6, XC = 15 mg/L FO, RC = 0.0023 min^{-1}	99%	[194]

CP = Chemical precipitation; PAN = polyacrylonitrile; MB = methylene blue; RhB = rhodamine B; MO = methylene orange; DR = direct red; RR = reactive red CBZ = carbamazepine, BPA = Bisphenol A; CR = Congo red (CR); Vis = visible; LI = light intensity; CT = calcination temperature; XC = xenobiotic concentration; CD = catalyst dose; CC = catalyst concentration; IT = irradiance time; PFO = pseudo first order kinetic model; FO = first order kinetic model; PC = phenol concentration; R&R = recycle and reuse; RR = reaction rate; RC = rate constant.

Table S9. CNTs-based membranes and filtration systems for wastewater treatment.

Membrane/Filter	Material	Methods	Membrane performance	Refs.
VACNT	CNT+PES	CVD	UF, WTS = 100 L/m². h at 60 Psi, 3 times faster than RO CNT+PES 10 times faster than PES membrane	[195]
VACNT	CNT+Epoxy	WAT CVD	3 times ⬆ WF than UF, 2 log ⬇ bacterial with VCNT than with UF PFR (after 600 min) = 67% for VCNT and 55% for UF	[196]
VACNT	CNT+SSM	Thermal CVD	Separate water layer and diesel and even SSE ⬆Hydrophobicity and oleophobicity	[197]
SWCNT	CNT+PVF	Vacuum filtration	⬇5×10^5 *E coli*, CI = 79% after 20 min CT, MA *E. Coli* = 6%, damaged *E coli* membrane	[198]
VACNT	CNT+PTE+Si *	WA CVD	Millimeter thick UFM, WP = 30000$Lm^{-2}h^{-1}bar\text{-}^{1}$, ⬇bacterial growth, ⬇biofilm formation	[199]
VACNT	MWCNT+Fe+Al_2O_3+Si *	Thermal CVD	PIF/TR = 24% (UFM), 69% (VCNTM), PRFR = 38% (UFM), 5% (VCNTM) in BSA ⬆ Removal 71% to 90% after surface modification by GP of MAA	[200]
VA DWCNT	DWCNT+Si wafer *	CVD	NaCl rejection = 41.4%, WF = 1.31×10^{-3} to 62.7×10^{-3} L cm^{-2} day^{-1} Mpa^{-1}	[201]
CNT BP	CNT	CVD, UV/OT/AS	SR ≥95%, LS ⬆ by 50%	[202]
VACNT	MCNT+PDMS+Si *	CVD	SR = 96.5% at OP of 2 bar	[203]
GO-VACNT PAC-VACNT	CNT+Epoxy	N.A.	SR = 45% (GO-VCNT), 65% (PAC-VCNT) at OP of 15.5 bar	[204]
PA/outer-wall VACNT	CNT+Epoxy	IP	ROM, Flux = 128.6 $Lm^{-2}h^{-1}$, SR = 98.3% at OP of 15.5 bar	[205]

N.A. = Not available; PES = poly ether sulfone; PVF = poly vinylidene fluoride; PTE = poly tetrafluoro ethylene; DWCNTs = double-wall CNT; PDMS = polydimethylsiloxane; SSM = stainless steel mesh; * substrate; VA = vertical aligned; VACNTs = vertical aligned CNTs; GO-VACNT = graphene oxide coated VACNT; PA polyamide; PAC-VACNT = polyamide coated VACNT; CNT BP = CNT bucky paper; IP = interfacial polymerization; CVD = chemical vapor deposition; WAT CVD = water assisted thermal CVD; WA CVD = water assisted CVD; OT = ozone treatment; AS = alkoxy silanation; UF = used for ultrafiltration; WTS = water transportation speed; RO CNT = random oriented CNT; WF = water flux; PFR = permeate flux reduction; SSE = surfactant stabilized emulsions; CI = cell inactivation, CT = contact time; MB = metabolically active; WP = water permeability; PIF/TR = proportion of irreversible fouling to total resistance; PRFR = proportion of reversible fouling resistance; BSA = bovine serum albumin; RI = removal improvement;

GP = graft-polymerization; MAA = methacrylic acid, WF = water flux; LS = life span; SR = salt rejection; OP = operational pressure; PO-VCNT = ; ROM = reverse osmosis membrane; UFM = ultrafiltration membrane; VCNTM = vertical CNT membrane; ↑ = high, higher, improve, improved.

Table S10. Applications of CNTs-based filtering membranes for water desalination.

Membrane	PM	OOP	Effect of CNTs	RE	Refs
MWCNT+CA	PI	1% CNT+CAM, FS = $MgSO_4$ WF =69.5 $Lm^{-2}h^{-1}$	↑Permeability	90.6% ($MgSO_4$), 83.3% (Na_2SO_4), 44.6% (NaCl)	[206]
CNT+PES	PI	CNT C = 0.1 wt%, WF =38.91$Lm^{-2}h^{-1}$ P = 4 bar; FS = 200 ppm Na_2SO_4, $MgSO_4$, NaCl	↑SR and WF	72.2% ($MgSO_4$), 87.3% (Na_2SO_4), 24.7% (NaCl)	[207]
PS20/BTCT/MPD/MWCNT	IP	FS = 2000 ppm NaCl, pH = 6–7, T= 25°C P = 225psi, WF = 43 $Lm^{-2}h^{-1}$	↑WP, WF, SH	99%	[208]
MWCNT-TN/PA	IP	P = 15 bar, WF = 0.74 $Lm^{-2}h^{-1}$ FS = 2000 ppm NaCl	↑CA, SC, SRGH	98% (NaCl), 98.1 (Na_2SO_4)	[209]
MVF PES/SPS/O-MWCNT	IP	FS = brackish waters; SC =2000 ppm, P= 3 bar WF = 30.2 $Lm^{-2}h^{-1}bar^{-1}$	↑SR, WP	SS (NaCl/Na_2SO_4) = 25	[210]
CNT/PA	E-A IP	FS = 1 g/L NaCl, P = 4 bar, WF = 96.8 $Lm^{-2}h^{-1}$	↑WF	89.6%	[211]
SWCNT/PA	Brush painting	FS = 1 g/L Na_2SO_4, P = 6 bar, WF = 40 $Lm^{-2}h^{-1}$	↑WP	96.5%	[212]
RGO/CNT	ED/CR	FS = 0.1 M NaCl, P = 1 bar, WF = 40.4±3.7 $Lm^{-2}h^{-1}$	↑WF	94±1.9%	[213]
PPC/MWCNT	IP	FS = 2000 mg/L Na_2SO_4, P = 1 MPa, = 65.7 $Lm^{-2}h^{-1}$	↑WP, SR	97.6 %	[214]

PM = Preparation methods; OOP = operation parameters; RE = removal efficiency; CA = cellulose acetate (CA); PES = poly ether sulfone; PS-20 = poly sulfone; BTCT = 1,3,5-benzenetricarbonyl trichloride; MPD = m-phenylenediamine; TN = titania nanotube; PAMVF = polyamide macro void-free PES; SPS = sulfonated poly sulfone; O-MWCNTs = oxidized MWCNT; RGO = reduced graphene oxide; PPC = poly pyrrole coated; IP = interfacial polymerization; E-A = electrospray-assisted; PI = phase inversion; ED = electrophoretic deposition; CR = chemical reduction; CAM = CA membrane; FS = feed system or feed solution; WF = water flux; CNT C = CNT concentration; P = pressure; T = temperature; SC = salt concentration; SS = salt selectivity; SR = salt rejection; WP = water permeability; SH = surface hydrophilicity; CA = contact angle; SC = surface charge; SRGH = surface roughness.

Table S11. Main developed methods for CNTs regeneration.

Adsorbents	Xenobiotic	Regeneration mode	Cycles	Results after last cycle	Ref.
O-MWCNT	Cadmium	Physical	-	At pH 1.5, 93% of Cd^{2+} RE	[215]
CNTs/ Fe_3O_4	Biphenol A	Chemical	5	No valuable loss in AC	[216]
Chitosan-MWCNTs	Congo Red dye	Chemical (0.01 M NaOH)	3	RR 71% after 3 cycles	[217]
O-MWCNT	BTX	Chemical	5	Only 5.15%, 0.97%, 1.05% (BTX) RE	[218]
MWCNTs- Cu-$NiFe_2O_4$	Oxytetracycline HCl	Chemical (0.1 M NaOH)	3	RE = 45.6%	[166]
Magnetic MWCNTs	Gatifloxacin	Chemical	5	Less than 7.8% recovery loss	[219]
MWCNTs	CO_2	TT and VI	20	Only 3% attrition after 20 cycles	[220]
MWCNTs	Anticancer drugs	TT	5	No negative influence on the sorption level	[221]
Magnetic MWCNTs	Atrazine	Ozone-assisted	10	Retained 85-93% of its AC	[171]
NaOCl-MWCNTs	Molybdenum	Chemical (0.15 M HNO_3)	10	89.5% RE with 3.6 wt% NaOCl	[222]
MWCNTs	AAP, ibuprofen, TCS	Chemical, ultrasonic and thermal	4-5	Sonication ⬆ RE%, 100% RE after TT (380 °C)	[223]
MWCNTs	Tetracycline	Microwave-UV system	5	AC of regenerated CNT = 100%	[224]
MWCNTs	Reactive red 3BS	Microwave irradiation	4	RE 92.8% but ⬇ AC	[225]
CNTs	Reactive Black dye	Electrochemical	2	RE 86.5% (MWCNTs) and 77.3% (SWCNTs)	[226]
Magnetic MWCNTs	p-Nitrophenol	Microwave-assisted	6	AC = 19.7 mg/g at 850 W, RE = 106%	[227]

AAP = Acetaminophen; TCS = triclosan; VI = Vacuum interactions; AC = absorption capacity; RE = regeneration efficiency; TT = thermal treatment; BTX = benzene, toluene and xylenes; RR = regeneration rate; UV = ultraviolet.

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