Supplementary Information

In-Situ Carbon Recovery from Refractory Organics in Wastewater: A Critical Review

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Table S1 Summary of representative systems for *in situ* carbon resource recovery of refractory organics

Treatment technology	Contaminant	Conditions	Degradation efficiency (%)	Product	Productivity	Selectivity （%）	Refs.
Photocatalysis	Carbamazepine (CBZ)	[CBZ]=0.02 g/L, [$BiOBr/BiVO_4$]=0.30 g/L, [NaOH]=0.01 M, wavelength range 350 nm ≤ λ ≤ 780 nm, light intensity 160 mW/cm^2, t=4 h, T=6°C	86.12	CO	2.56 $\mu mol\cdot g^{-1}\cdot h^{-1}$	100	1
	Methylene Blue (MB)	[MB]=0.008 g/L, [5%GQDs/V-TiO_2]=1.00 g/L, wavelength range 320 nm ≤ λ ≤ 780 nm, light intensity 160 mW/cm^2, t=8 h, T=6°C	99	CH_3OH C_2H_5OH CH_4	13.24 $\mu mol\cdot g^{-1}\cdot h^{-1}$ 5.65 $\mu mol\cdot g^{-1}\cdot h^{-1}$ 0.445 $\mu mol\cdot g^{-1}\cdot h^{-1}$	68.5 29.2 2.30	2
	Rhodamine (RhB)	[RhB]=0.2 mM, [1%rGO/$SrTi_{0.95}Fe_{0.05}O_{3-\sigma}$]=1.00 g/L, wavelength range 320 nm ≤ λ ≤ 780 nm, light intensity 160 mW/cm^2, t=6 h, T=6°C	90	CH_3OH C_2H_5OH	10.76 $\mu mol\cdot g^{-1}\cdot h^{-1}$ 6.14 $\mu mol\cdot g^{-1}\cdot h^{-1}$	63.7 36.3	3
	RhB	[RhB]=0.01 g/L, [$Ti_3C_2T_x/TiO_2$]=0.5 g/L, wavelength range 320 nm ≤ λ ≤ 780 nm, light intensity 160 mW/cm^2, t=1 h	77.60	CO	10.02 $\mu mol\cdot g^{-1}\cdot h^{-1}$	/	4
	Phenol	[Phenol]=0.02 g/L, [Co/In_2O_3-550]=0.2 g/L, [[$Ru(bpy)_3]Cl_2\cdot 6H_2O$]=0.2 g/L, wavelength range 200 nm≤λ≤1200 nm, light intensity 100 mW/cm^2, t=8 h, T=25°C	30	CO	0.9 $\mu mol\cdot g^{-1}\cdot h^{-1}$	/	5
	RhB	[RhB]=0.01 g/L, [g-C_3N_4/Bi/$BiVO_4$]=1.00 g/L, wavelength range 420 nm ≤ λ ≤ 780 nm, light intensity 160 mW/cm^2, t=1 h	30	CO	0.63 $\mu mol\cdot g^{-1}\cdot h^{-1}$	/	6
Electrocatalysis -AOPs	4-nitrophenol (4-NP)	[4-NP]=0.01 g/L, [PMS]=0.5 g/L, anode=Co0.5, cathode=flower-like CuO, U=0.8 V, Na_2SO_4 (anolyte) = 0.1 M, $KHCO_3$ (catholyte) = 0.1 M, t=2 h, T=25°C	99	CH_3OH C_2H_5OH	49.15 $\mu mol\cdot L^{-1}\cdot h^{-1}$ 20.48 $\mu mol\cdot L^{-1}\cdot h^{-1}$	65.7 27.4	7
	Salicylic acid (SA)	[SA]=0.05 g/L, [PS]=0.02M, anode=MWCNT-CF, cathode=sFe-Cu@MOF, I = 0.025 A, [Na_2SO_4 (anolyte)]=0.1 M, [$KHCO_3$ (catholyte)]=0.1 M, t=3 h, T=25°C	99.1±0.9	HCOOH	8.00 $\mu mol\cdot L^{-1}\cdot h^{-1}$	/	8

Table S1 (*continued*)

	4-NP	[4-NP]=0.01 g/L, [PMS]=0.5 g/L, anode=Co0.5@C, cathode=SnO_2/CC, U=1.3 V, [Na_2SO_4 (anolyte)] = 0.1 M, [$KHCO_3$ (catholyte)] = 0.1 M, t=1 h, T=25℃	99.0	HCOOH	39.19 $\mu mol \cdot L^{-1} \cdot h^{-1}$	/	[9]
Photo-electrocatalysis	Phenol	[Phenol]=0.01 g/L, [Na_2SO_4]=0.1M, anode=Pd-TiO_2, cathode=Pd-Cu, wavelength range 200 nm≤λ≤1200 nm, light intensity 100 mW/cm^2, current density=10 mW/cm^2, t=8 h, T=25℃	87	CO	60.59 $\mu mol \cdot L^{-1} \cdot h^{-1}$	99.8	[10]
	Methyl orange (MO)	[MO]=0.02 g/L, anode=TiO_2-NT, cathode=Ti_3C_2, pH(anode)=7.0, pH(cathode)=1.0, wavelength range λ≤254 nm, light intensity 3.60 mW/cm^2，t=5 h, T=25℃	74.8	CH_4	5.2$\mu mol \cdot g^{-1} \cdot h^{-1}$	77.6	[11]
	Phenol	[Phenol]=0.01 g/L, [KOH]=0.5M, anode=Pd QDs@TiO_2 NTPC, cathode=Co_3O_4 NWs, wavelength range 200 nm≤λ≤1200 nm, light intensity 100 mW/cm^2, U=3.0V, t=8 h, T=25℃	84.7	CO	2.39 $\mu mol \cdot L^{-1} \cdot h^{-}$	5.2	[12]
				HCOOH	43.42 $\mu mol \cdot L^{-1} \cdot h^{-}$	94.8	
Piezocatalysis-AOPs	Phenol	[Phenol]=0.01 g/L, [PMS]=3 g/L, [Co_3S_4/MoS_2]=0.6 g/L, ultrasonic frequency=40 kHz, t=4 h, T=20℃	99.90	CO	17.33 $\mu mol \cdot g^{-1} \cdot h^{-1}$	74.5	[13]

Table S2 Cost assessment of each technology

Technology	Category	Item	Unit-price		Dosage		Total (USD)	Cost of category (USD)	Total TOC eliminated (mg)	Cost of category (USD/mg TOC)
			Value	Unit	Value	Unit				
Photocatalysis[1]	Electricity	Ultrasonic stirrer	0.07	USD/kW·h	0.0004	kW·h	0.00003	0.3503	0.8000	0.4378
		Drying oven	0.07	USD/kW·h	0.0032	kW·h	0.00022			
		Centrifugal machine	0.07	USD/kW·h	0.0002	kW·h	0.00001			
		Oil pump	0.07	USD/kW·h	0.4000	kW·h	0.02800			
		Cryogenic circulating pump	0.07	USD/kW·h	3.4000	kW·h	0.23800			
		Xe lamp	0.07	USD/kW·h	1.20000	kW·h	0.08400			
	Reagent	$Bi(NO_3)_3 \cdot 5H_2O$	0.44	USD/g	0.0116	g	0.00508	0.6988		0.8735
		Nitric acid	0.20	USD/mL	0.0600	ml	0.00985			
		NH_4VO_3	0.50	USD/g	0.0014	g	0.00072			
		KBr	0.10	USD/g	0.0017	g	0.00018			
		NaOH	1.71	USD/g	0.4000	g	0.68300			
Electrocatalysis-AOPs[7]	Electricity	Drying oven	0.07	USD/kW·h	4.8400	kW·h	0.33880	0.4236	0.4122	1.0277
		Tube furnace	0.07	USD/kW·h	1.2000	kW·h	0.08400			
		Ultrasonic stirrer	0.07	USD/kW·h	0.0100	kW·h	0.00070			
		Electrochemical reactor	0.07	USD/kW·h	0.0016	kW·h	0.00013			
	Reagent	$Co(NO_3)_2$ $6H_2O$	3.18	USD/g	0.0582	g	0.18510	2.9972		7.2713
		Ethylene glycol	0.04	USD/mL	1.0000	mL	0.04380			
		Urea	0.04	USD/g	0.0500	g	0.00209			
		NH_4F	0.16	USD/g	0.0185	g	0.00295			

Table S2 (*continued*)

Technology	Category	Item	Unit-price		Dosage		Total (USD)	Cost of category (USD)	Total TOC eliminated (mg)	Cost of category (USD/mg TOC)
			Value	Unit	Value	Unit				
		Cetyltrimethylammonium bromide	0.43	USD/g	0.0050	g	0.00215			
		Conductive glass	0.27	USD/g	0.1650	g	0.04469			
		NaOH	1.71	USD/g	0.6400	g	1.09280			
		$(NH_4)_2S_2O_8$	0.07	USD/g	0.1096	g	0.00787			
		$Na_2WO_4{\cdot}2H_2O$	0.67	USD/g	0.0211	g	0.01412			
		Sodium dodecyl sulfate	1.88	USD/g	0.0461	g	0.08656			
		Copper mesh	1.60	USD/g	0.0669	g	0.10682			
		Acetone	0.06	USD/mL	10.0000	mL	0.55947			
		Ethanol	0.04	USD/mL	10.0000	mL	0.43943			
		Na_2SO_4	0.43	USD/g	0.8520	g	0.36960			
		$KHCO_3$	0.06	USD/g	0.6010	g	0.03934			
		PMS	0.24	USD/g	0.0060	g	0.00143			
		Ultrasonic stirrer	0.07	USD/kW·h	0.0107	kW·h	0.00075			
		Drying oven	0.07	USD/kW·h	2.5950	kW·h	0.18165			
		Tube furnace	0.07	USD/kW·h	0.3000	kW·h	0.02100			
	Electricity	Electrochemical reactor	0.07	USD/kW·h	0.0040	kW·h	0.00028	0.4278		0.6470
Photo-electrocatalysis[10]		Magnetic stirrers	0.07	USD/kW·h	0.4002	kW·h	0.02801		0.6612	
		Oil pump	0.07	USD/kW·h	0.4000	kW·h	0.00280			
		Xe lamp	0.07	USD/kW·h	2.4000	kW·h	0.16800			
	Reagent	Ti	1.47	USD/g	0.5063	g	0.74630	8.3275		12.5946
		Acetone	0.06	USD/mL	10.0000	mL	0.55947			

Table S2 (*continued*)

Technology	Category	Item	Unit-price	Dosage	Total	Cost of	Total TOC	Cost of category

			Value	Unit	Value	Unit	(USD)	category (USD)	eliminated (mg)	(USD/mg TOC)
		Ethanol	0.04	USD/ml	10.0000	mL	0.43943			
		Ethylene glycol	0.04	USD/ml	10.000	mL	0.43800			
		NH_4F	0.16	USD/g	0.0333	g	0.00531			
		Polyvinylpyrrolidone	1.19	USD/g	0.0800	g	0.09512			
		$PdCl_2$	739.90	USD/g	0.0009	g	0.67375			
		NaI	0.58	USD/g	0.0300	g	0.01727			
		Copper mesh	1.60	USD/g	0.0669	g	0.10682			
		HCl	0.39	USD/mL	6.0000	mL	2.33958			
		H_3PO_4	0.38	USD/mL	6.0000	mL	2.29023			
		Na_2SO_4	0.43	USD/g	1.4205	g	0.61621			
		Magnetic stirrers	0.07	USD/kW·h	0.0002	kW·h	0.00001			
		Drying oven	0.07	USD/kW·h	0.8060	kW·h	0.05642			
	Electricity	Water bath stirrer	0.07	USD/kW·h	0.0022	kW·h	0.00015	0.4346		1.5188
		Oil pump	0.07	USD/kW·h	0.4000	kW·h	0.02800			
		Ultrasonic generator	0.07	USD/kW·h	1.6000	kW·h	0.11200			
Piezocatalysis-AOPs[13]		Cryogenic circulating pump	0.07	USD/kW·h	3.4000	kW·h	0.23800		0.2861	
		$H_{24}Mo_7N_6O_{24}\cdot 4H_2O$	0.40	USD/g	0.0120	g	0.00475			
		C_2H_5NS	2.36	USD/g	0.0085	g	0.02003			
	Reagent	$Co(NO_3)_2\cdot 6H_2O$	3.18	USD/g	0.0040	g	0.01272	0.0746		0.2608
		$H_4N_2\cdot H_2O$	0.57	USD/mL	0.0023	mL	0.00129			
		PMS	0.24	USD/g	0.1500	g	0.03585			

Table S3 Cost of each technology for *in situ* utilization of carbon resource

Technologies	$Cost_{EE}$ (\$/mg TOC)	$Cost_{Reagent}$ (\$/mg TOC)	$Cost_{Total}$ (\$/mg TOC)
Photocatalysis	0.44	0.87	1.31
Electrocatalysis-AOPs	1.03	7.27	8.30
Photo-electrocatalysis	0.65	12.59	13.24
Piezocatalysis-AOPs	1.52	0.26	1.78

Table S4. Life cycle inventory for each technology normalized to the functional unit.

Technology	Stage	Category	Item	Value	Unit
Photocatalysis[1]	Catalyst preparation	Input	$Bi(NO_3)_3 \cdot 5H_2O$	0.0145	g
			Nitric acid	0.0048	g
			Ultrapure water	1.5759	g
			NH_4VO_3	0.0018	g
			KBr	0.0021	g
			Ultrasonic stirrer	0.0005	kw·h
			Drying oven	0.0040	kw·h
			Centrifugal machine	0.0003	kw·h
		Output	Waste liquid discharge	1.5898	g
	Catalytic reaction	Input	CBZ	0.0025	g
			Ultrapure water	125.0000	g
			NaOH	0.5000	g
			Ar	133.8000	g
			Xe lamp	1.5000	kw·h
			Cryogenic circulating	4.2500	kw·h
			Oil pump	0.5000	kw·h
		Output	Waste liquid discharge	125.4969	g
			Emission to air:Ar	133.8000	g
Electrocatalysis-AOPs[7]	Catalyst preparation	Input	$Co(NO_3)_2$ $6H_2O$	0.1412	g
			Ethylene glycol	2.7001	g
			Urea	0.1213	g
			NH_4F	0.0449	g
			Cetyltrimethylammonium bromide	0.0121	g
			Conductive glass	0.4003	g
			NaOH	1.5526	g
			$(NH_4)_2S_2O_8$	0.2659	g
			$Na_2WO_4 \cdot 2H_2O$	0.0512	g
			Sodium dodecyl sulfate	0.1118	g
			Copper mesh	0.1623	g
			Acetone	19.1897	g

Table S4 (*continued*)

Technology	Stage	Category	Item	Value	Unit
			Ethanol	19.1412	g
			Ultrapure water	184.8617	g
		Output	Waste liquid discharge	228.5143	g
			4-NP	0.0015	g
			Na_2SO_4	2.0670	g
	Catalytic reaction	Input	$KHCO_3$	1.4580	g
			PMS	0.0146	g
			Electrochemical reactor	0.0039	kw·h
		Output	Waste liquid discharge	294.6616	g
			Ti	0.7657	g
			Acetone	11.9631	g
			Ethanol	11.9328	g
			Ethylene glycol	16.4398	g
			NH_4F	0.0504	g
			Polyvinylpyrrolidone	0.1210	g
			$PdCl_2$	0.0014	g
			NaI	0.0454	g
	Catalyst preparation	Input	Copper mesh	0.1012	g
			HCl	0.5522	g
			H_3PO_4	13.2184	g
Photo-electrocatalysis[10]			Ultrapure water	201.1494	g
			Ultrasonic stirrer	0.0162	kw·h
			Drying oven	3.9247	kw·h
			Tube furnace	0.4537	kw·h
			Electrochemical reactor	0.0042	kw·h
			Magnetic stirrers	0.0003	kw·h
		Output	Waste liquid discharge	266.9239	g
			Na_2SO_4	2.1484	g
			Phenol	0.0015	g
	Catalytic reaction	Input	N_2	3.3727	g
			Ultrapure water	151.2402	g
			Oil pump	0.6050	kw·h
			Electrochemical reactor	0.0018	kw·h

Table S4 (*continued*)

Technology	Stage	Category	Item	Value	Unit
		Output	Xe lamp	3.6298	kw·h
			Magnetic stirrers	0.6050	kw·h
			Waste liquid discharge	153.3878	g
			Emission to air: CO	0.0023	g
			Emission to air: N_2	3.3727	g
Piezocatalysis-AOPs[13]	Catalyst preparation	Input	$H_{24}Mo_7N_6O_{24}\cdot 4H_2O$	0.0419	g
			C_2H_5NS	0.0297	g
			$Co(NO_3)_2\cdot 6H_2O$	0.0140	g
			$H_4N_2\cdot H_2O$	0.0783	g
			Ultrapure water	25.8235	g
			Magnetic stirrers	0.0007	kw·h
			Drying oven	2.8172	kw·h
			Water bath stirrer	0.0077	kw·h
		Output	Waste liquid discharge	25.9567	g
			Emission to air: NH_3	0.0021	g
			Emission to air: SO_2	0.0024	g
	Catalytic reaction	Input	PMS	0.5243	g
			Ar	374.1349	g
			Phenol	0.0017	g
			Ultrapure water	174.7641	g
			Oil pump	1.3981	kw·h
			Ultrasonic generator	5.5925	kw·h
			Cryogenic circulating pump	11.8840	kw·h
		Output	Waste liquid discharge	175.2898	g
			Emission to air:Ar	374.1349	g

Table S5. Results for the different systems/stages per functional unit (CML-IA baseline (World 2000))

Impact category	Unit	Photocatalysis			Electrocatalysis			Photo-electrocatalysis			Piezocatalysis		
		Total	Preparation	Reaction	Total	Preparation	Reaction	Total	Preparation	Reaction	Total	Preparation	Reaction
Abiotic depletion	kg Sb eq	3.51×10^{-6}	1.26×10^{-7}	3.39×10^{-6}	2.33×10^{-5}	2.33×10^{-5}	2.22×10^{-9}	5.79×10^{-6}	3.22×10^{-6}	2.57×10^{-6}	1.06×10^{-5}	1.64×10^{-6}	8.99×10^{-6}
Abiotic depletion (fossil fuels)	MJ	5.57×10^{1}	3.29	5.24×10^{1}	1.32×10^{2}	1.32×10^{2}	3.43×10^{-2}	7.85×10^{1}	3.88×10^{1}	3.97×10^{1}	1.46×10^{2}	2.56×10^{1}	1.21×10^{2}
Global warming (GWP 100a)	kg CO_2 eq	6.22	3.01×10^{-1}	5.92	1.48×10^{1}	1.48×10^{1}	3.88×10^{-3}	8.71	4.22	4.49	1.34×10^{1}	2.35	1.11×10^{1}
Ozone layer depletion (ODP)	kg CFC-11 eq	6.35×10^{-8}	1.52×10^{-8}	4.83×10^{-8}	1.28×10^{-7}	1.28×10^{-7}	3.16E-11	7.90×10^{-8}	4.24×10^{-8}	3.66×10^{-8}	6.38×10^{-7}	1.14×10^{-7}	5.24×10^{-7}
Human toxicity	kg 1,4-DB eq	2.21	1.14×10^{-1}	2.10	5.32	5.32	1.37×10^{-3}	3.14	1.55	1.59	5.45	9.37×10^{-1}	4.51
Fresh water aquatic ecotoxicity	kg 1,4-DB eq	2.47	1.23×10^{-1}	2.35	5.88	5.87	1.54×10^{-3}	3.47	1.69	1.78	7.81	1.26	6.55
Marine aquatic ecotoxicity	kg 1,4-DB eq	1.00×10^{4}	4.68×10^{2}	9.55×10^{3}	2.38×10^{4}	2.38×10^{4}	6.25	1.40×10^{4}	6.76×10^{3}	7.24×10^{3}	2.19×10^{4}	3.79×10^{3}	1.81×10^{4}
Terrestrial ecotoxicity	kg 1,4-DB eq	2.27×10^{-2}	6.03×10^{-4}	2.21×10^{-2}	5.52×10^{-2}	5.52×10^{-2}	1.45×10^{-5}	3.23×10^{-2}	1.56×10^{-2}	1.68×10^{-2}	6.33×10^{-2}	9.39×10^{-3}	5.39×10^{-2}
Photochemical oxidation	kg C_2H_4 eq	1.04×10^{-3}	5.17×10^{-5}	9.89×10^{-4}	2.52×10^{-3}	2.52×10^{-3}	6.47×10^{-7}	1.51×10^{-3}	7.61×10^{-4}	7.50×10^{-4}	2.34×10^{-3}	4.07×10^{-4}	1.93×10^{-3}
Acidification	kg SO_2 eq	2.78×10^{-2}	1.31×10^{-3}	2.64×10^{-2}	6.62×10^{-2}	6.62×10^{-2}	1.73×10^{-5}	3.91×10^{-2}	1.91×10^{-2}	2.00×10^{-2}	5.91×10^{-2}	1.03×10^{-2}	4.88×10^{-2}
Eutrophication	kg PO_4^{3-} eq	6.69×10^{-3}	6.35×10^{-4}	6.05×10^{-3}	1.53×10^{-2}	1.53×10^{-2}	3.96×10^{-6}	9.10×10^{-3}	4.52×10^{-3}	4.59×10^{-3}	2.83×10^{-2}	4.94×10^{-3}	2.34×10^{-2}

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