

Supplementary materials

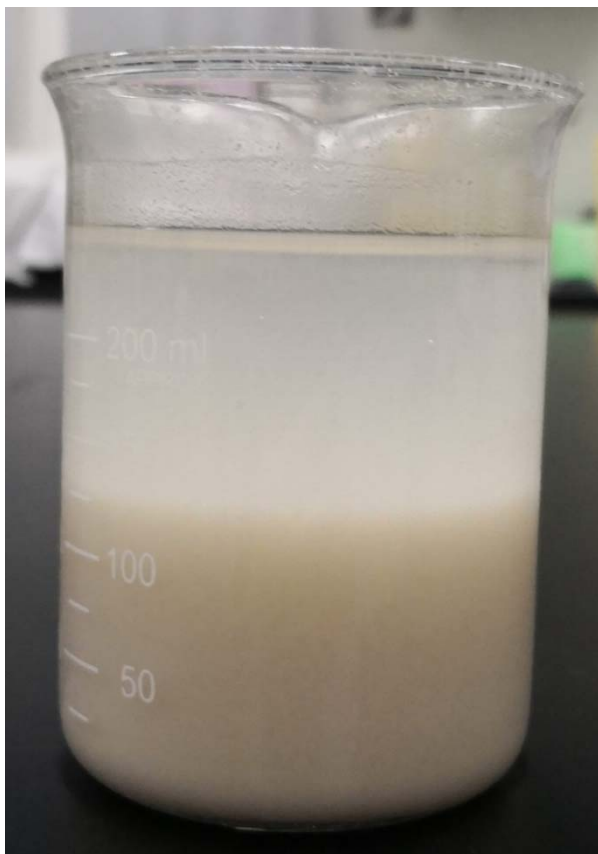


Fig. S1 The Mn^0 materials synthesis via the borohydride reduction method

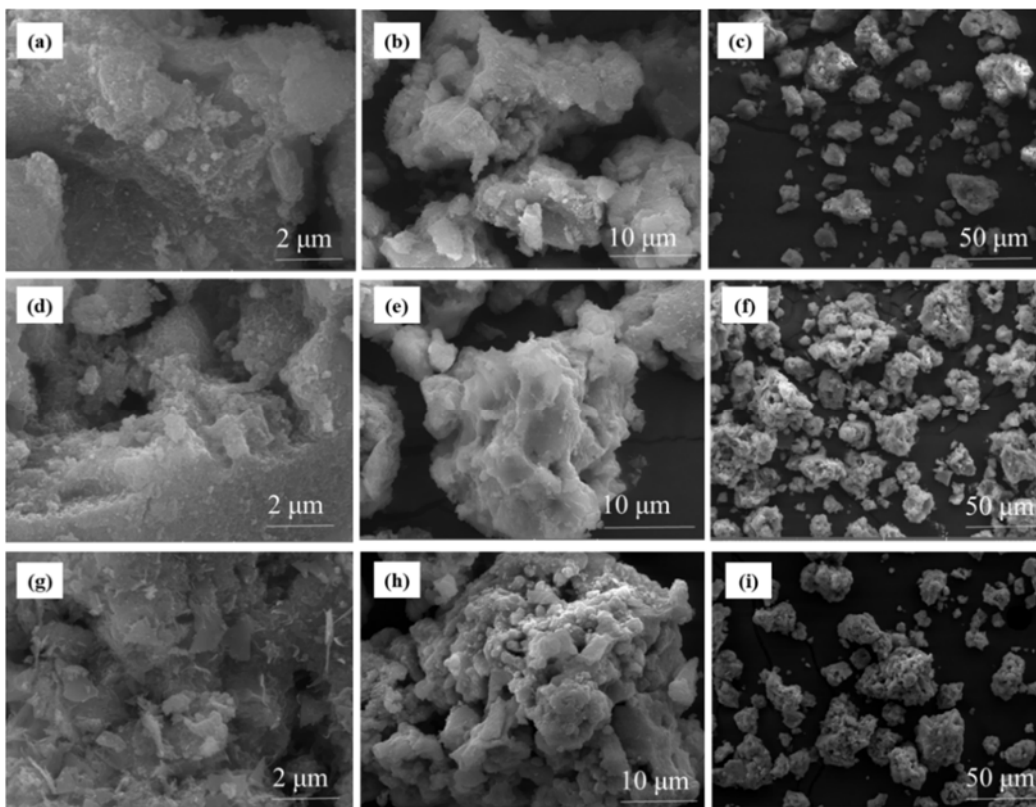


Fig. S2 SEM images of the Mn⁰: (a), (b) and (c) before Tl(I) removal; (d), (e) and (f) after removal at Tl(I)₀ = 20 mg/L; (g), (h) and (i) after removal at Tl(I)₀ = 200 mg/L

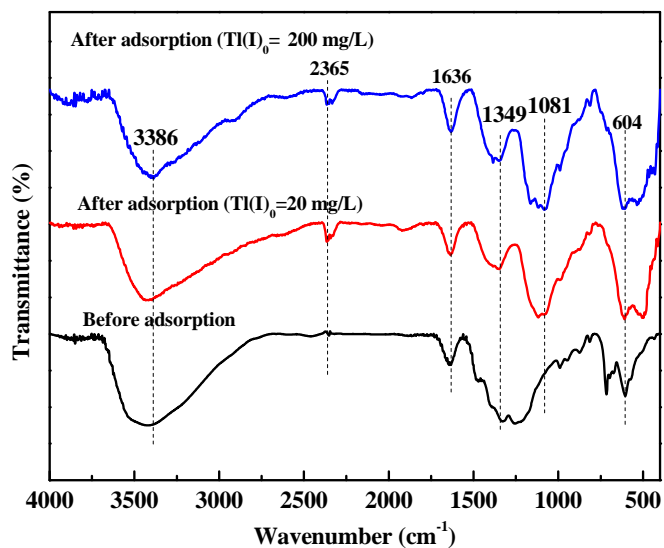
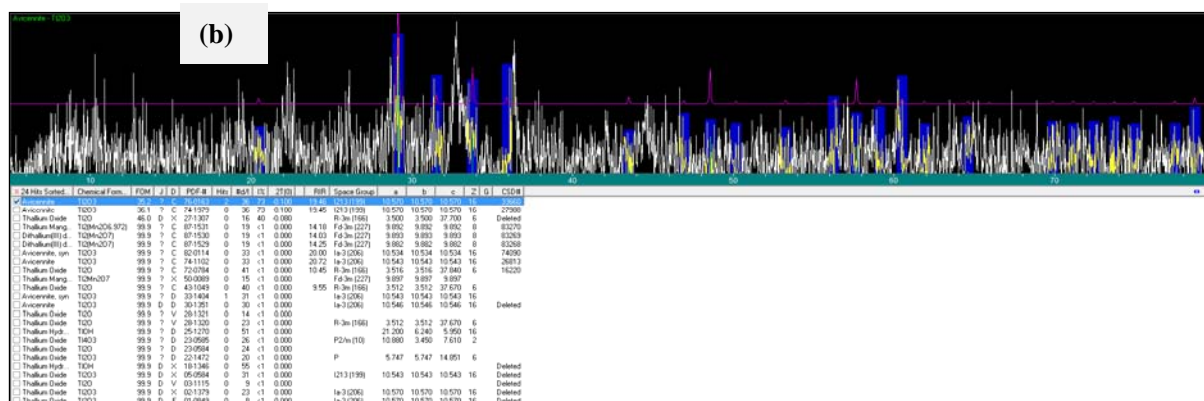
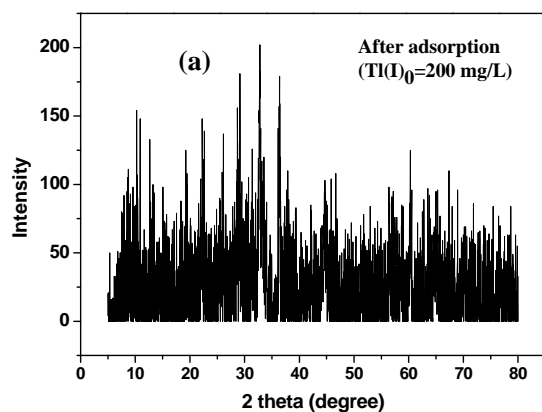


Fig. S3 FT-IR spectra of the Mn⁰ before and after oxidation treatment with persulfate for Tl(I) removal



Chemical Form...	FQM	J	D	PDF-#	Hits	#d/l	I%	2T(0)	RIR	Space Group	a	b	c	Z	G	CSD#
<input checked="" type="checkbox"/> Avicennite	Tl2O3	35.2	?	C	76-0163	2	36	73	-0.100	19.46	I213 (199)	10.570	10.570	10.570	16	33660
<input type="checkbox"/> Avicennite	Tl2O3	36.1	?	C	74-1979	0	36	73	-0.100	19.45	I213 (199)	10.570	10.570	10.570	16	27988
<input type="checkbox"/> Thallium Oxide	Tl2O	46.0	D	X	27-1307	0	16	40	-0.080		R-3m (166)	3.500	3.500	37.700	6	Deleted
<input type="checkbox"/> Thallium Mang...	Tl2(Mn2O6.972)	99.9	?	C	87-1531	0	19	<1	0.000	14.18	Fd-3m (227)	9.892	9.892	9.892	8	83270
<input type="checkbox"/> Dithallium(III) d...	Tl2(Mn2O7)	99.9	?	C	87-1530	0	19	<1	0.000	14.03	Fd-3m (227)	9.893	9.893	9.893	8	83269
<input type="checkbox"/> Dithallium(III) d...	Tl2(Mn2O7)	99.9	?	C	87-1529	0	19	<1	0.000	14.25	Fd-3m (227)	9.882	9.882	9.882	8	83268
<input type="checkbox"/> Avicennite, syn	Tl2O3	99.9	?	C	82-0114	0	33	<1	0.000	20.00	la-3 (206)	10.534	10.534	10.534	16	74090
<input type="checkbox"/> Avicennite	Tl2O3	99.9	?	C	74-1102	0	33	<1	0.000	20.72	la-3 (206)	10.543	10.543	10.543	16	26813
<input type="checkbox"/> Thallium Oxide	Tl2O	99.9	?	C	72-0784	0	41	<1	0.000	10.45	R-3m (166)	3.516	3.516	37.840	6	16220
<input type="checkbox"/> Thallium Mang...	Tl2Mn2O7	99.9	?	X	50-0089	0	15	<1	0.000		Fd-3m (227)	9.897	9.897	9.897		
<input type="checkbox"/> Thallium Oxide	Tl2O	99.9	?	C	43-1049	0	40	<1	0.000	9.55	R-3m (166)	3.512	3.512	37.670	6	
<input type="checkbox"/> Avicennite, syn	Tl2O3	99.9	?	D	33-1404	1	31	<1	0.000		la-3 (206)	10.543	10.543	10.543	16	
<input type="checkbox"/> Avicennite	Tl2O3	99.9	?	D	30-1351	0	30	<1	0.000		la-3 (206)	10.546	10.546	10.546	16	Deleted
<input type="checkbox"/> Thallium Oxide	Tl2O	?	?	V	28-1321	0	14	<1	0.000							
<input type="checkbox"/> Thallium Oxide	Tl2O	?	?	V	28-1320	0	23	<1	0.000		R-3m (166)	3.512	3.512	37.670	6	
<input type="checkbox"/> Thallium Hydr...	TlOH	99.9	?	D	25-1270	0	51	<1	0.000			21.200	6.240	5.950	16	
<input type="checkbox"/> Thallium Oxide	Tl4O3	99.9	?	D	23-0585	0	26	<1	0.000		P2/m (10)	10.880	3.450	7.610	2	
<input type="checkbox"/> Thallium Oxide	Tl2O	99.9	?	D	23-0584	0	24	<1	0.000							
<input type="checkbox"/> Thallium Oxide	Tl2O3	99.9	?	D	22-1472	0	20	<1	0.000		P	5.747	5.747	14.851	6	
<input type="checkbox"/> Thallium Hydr...	TlOH	99.9	D	X	18-1346	0	55	<1	0.000							Deleted
<input type="checkbox"/> Thallium Oxide	Tl2O3	99.9	D	X	05-0584	0	31	<1	0.000		I213 (199)	10.543	10.543	10.543	16	Deleted
<input type="checkbox"/> Thallium Oxide	Tl2O	99.9	D	V	03-1115	0	9	<1	0.000							Deleted
<input type="checkbox"/> Thallium Oxide	Tl2O3	99.9	D	X	02-1379	0	23	<1	0.000		la-3 (206)	10.570	10.570	10.570	16	Deleted
<input type="checkbox"/> Thallium Oxide	Tl2O3	99.9	D	F	01-0849	0	8	<1	0.000		la-3 (206)	10.570	10.570	10.570	16	Deleted

Fig. S4 The XRD patterns of the Mn⁰ after adsorption of Tl(I)₀ = 200 mg/L: (a) profiles with elimination of background, (b) retrieval with software Jade 6.0, (c) the magnified retrieval list with software Jade 6.0 showing the avicennite (Tl₂O₃) listed in the 1st place

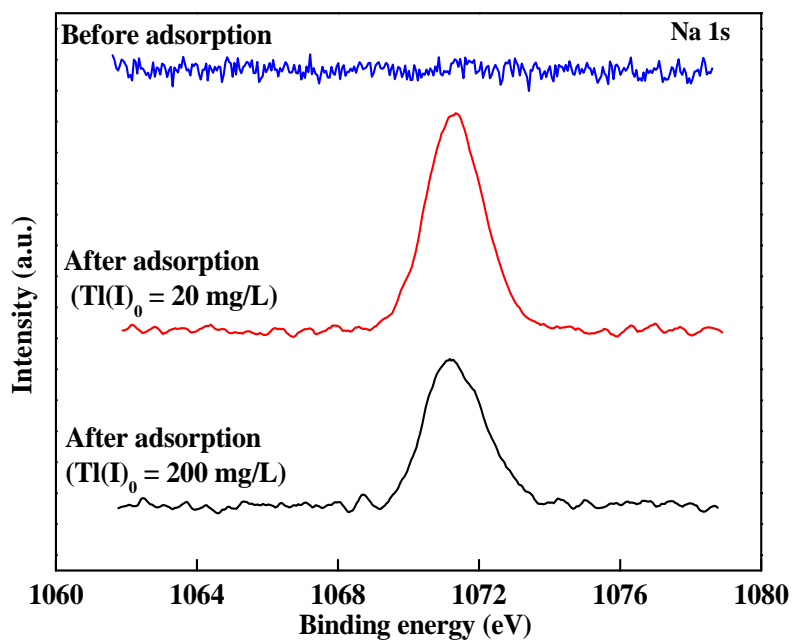


Fig. S5 XPS spectra of the Mn⁰ before and after adsorption of Tl(I): Na 1s core level

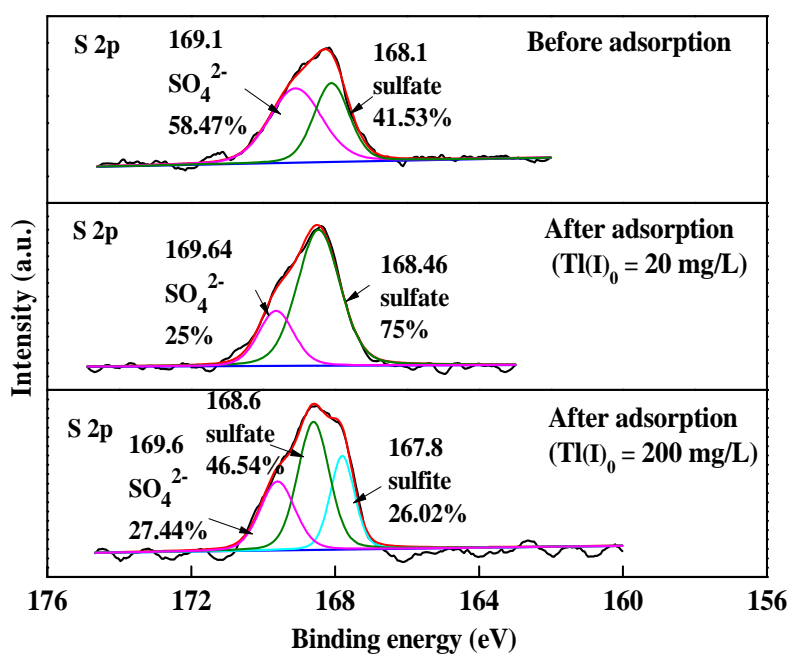


Fig. S6 XPS spectra of the Mn⁰ before and after adsorption of Tl(I): S 2p core level

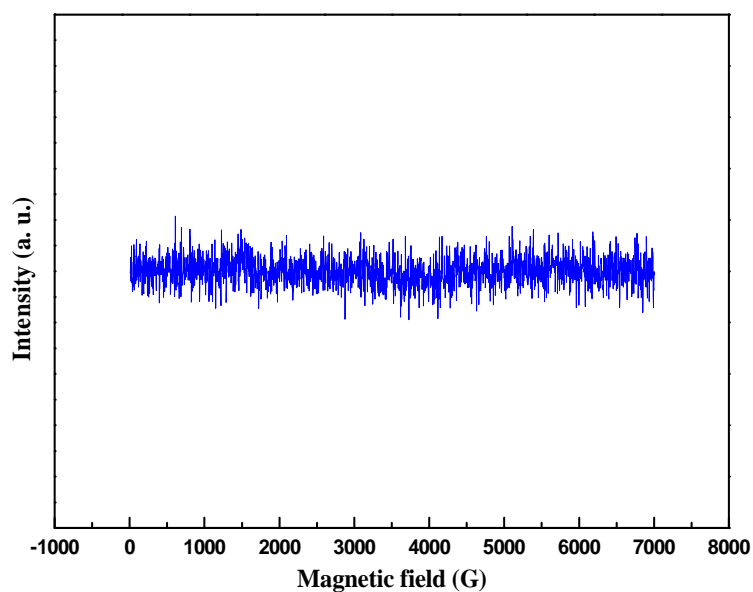


Fig. S7 ESR spectra of the free oxygenous radicals captured by DMPO: the control group with oxidant addition alone

Table S1 EDS-based elemental composition of the adsorbent before and after adsorption

Elements	Wt%			At%		
	Tl(I) ₀ = 0	Tl(I) ₀ = 20	Tl(I) ₀ = 200	Tl(I) ₀ = 0	Tl(I) ₀ = 20	Tl(I) ₀ = 200
	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L
C	7.55	8.36	8.97	14.1	16.3	18.4
N	0.950	0.796	0.905	1.52	1.34	1.59
O	46.3	39.8	35.2	64.8	58.4	54.3
Na	0.386	3.23	3.01	0.384	3.29	3.24
S	3.05	2.51	3.12	2.13	1.84	2.40
Mn	41.8	43.7	43.2	17.0	18.6	19.4
Tl	0.00	1.64	5.59	0.00	0.194	0.674

Table S2 The solution pH after reaction during experiments of effects of initial pH on Tl removal

Initial pH	The solution pH after reaction		
	Na ₂ S ₂ O ₈	H ₂ O ₂	NaClO
2	7.0	7.5	3.3
4	7.1	7.8	7.0
6	7.1	7.5	7.3
8	7.1	7.5	7.2
10	7.0	7.8	7.4
12	8.8	11.4	11.2

Table S3 Langmuir, Freundlich, and Temkin isotherm parameters for Tl(I) removal using $\text{Mn}^0\text{-S}_2\text{O}_8^{2-}$ system at pH 10

Langmuir model			Freundlich model			Temkin model		
Q_m (mg/g)	K_L (L/mg)	R^2	K_F (L/g)	$1/n$	R^2	A (L/g)	b (KJ/mol)	R^2
460	0.0100	0.942	14.3	0.589	0.943	0.423	0.0346	0.824

Table S4 Comparison of Tl(I) removal capacity via $\text{Mn}^0/\text{S}_2\text{O}_8^{2-}$ system with reported adsorbents

Adsorbents	Adsorbent dosage (g/L)	Tl equilibrium concentration (mg/L)	Reaction time (h)	Max. Adsorption capacity (mg/g)	References
Nano-hydroous MnO_2	0.5	0–100	24	353.6	Wan et al. (2014)
FeOOH-loaded MnO_2	0.25	0–700	24	236.4	Chen et al. (2017)
Nano- Al_2O_3	3.0	0–35	0.017	4.8	Zhang et al. (2008)
Titanium peroxide	0.2	0–50	24	412	Zhang et al. (2018)
Titanate nanotubes	0.2	0–60	3	709.2	Liu et al. (2014)
Sawdust	100	0–1000	24	13.2	Memon et al. (2008)
Sugar beet pulp	7	0–20000	0.25	185.2	Zolgharnein et al. (2011)
Alginate-Prussian blue	1	0–400	72	103	Vincent et al. (2014)
Fe_3O_4	2.5	0–500	0.5	104.8	Li et al. (2018)
Carbon nanotube	0.1	0–0.12	24	0.42	Rehman et al. (2012)
PAAm-Z	10	0–1000	24	378.1	Senol and Ulusoy, (2010)
Mn^0	2	10–500	0.5	460.1	This study

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