Electronic Supplementary Materials

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Effective removal of Sb(V) from aqueous solutions by micro-electrolysis with composite scrap iron-manganese as filler

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Table S1 Composition and concentration of artificially simulated wastewater

Reagent	Concentration(mg/L)	Reagent	Concentration(mg/L)
$C_6H_{12}O_6$	1000	Na_2SO_4	800
NH_4Cl	80	$NaHCO_3$	60
KH_2PO_4	20	$MgSO_4 \cdot 7H_2O$	30
Sb(V)	1000	$Na_2S \cdot 9H_2O$	400

Table S2 Designed working conditions

Stage	No.	HRT(h)	Mn mass ratio	Fe/C mass ratio
I [a]	HRT-8	8	0%	1.6 : 1
		8	20 %	1.6 : 1
	HRT-10	10	0 %	1.6 : 1
		10	20%	1.6 : 1
	HRT-12	12	0 %	1.6 : 1
		12	20%	1.6 : 1
	HRT-16	16	0 %	1.6 : 1
		16	20%	1.6 : 1
	HRT-24	24	0 %	1.6 : 1
		24	20%	1.6 : 1
II [b]	Fe-2C	24	10%	1 : 2
	Fe-C	24	10%	1 : 1
	2Fe-C [c]	24	10%	2 : 1
	3Fe-C	24	10%	3 : 1
	0%-Mn	24	0%	2 : 1
	11.11%-Mn	24	11.11%	2 : 1
	14.29%-Mn	24	14.29%	2 : 1
	25.00%-Mn	24	25%	2 : 1

a. Stage I is focused on investigating the impact of hydraulic retention time (HRT).
b. Stage II aims to examine the effects of varying Fe/C/Mn ratios.
c. The condition can also be named as 10%-Mn to investigate the variation of Sb(V) removal by Fe-Mn-C ME with different Mn mass ratio.

Table S3 Specific surface area of Fe-Mn flocs and Fe flocs

No.	Mass ratio of Mn	BET specific surface area (m^2/g)
0%-Mn	0%	117.67
14.29%-Mn	14.29%	130.79
25.00%-Mn	25.00%	121.36

Table S4 Fe(II) and Fe(III) content of Fe-Mn flocs at different Mn mass ratios

No.	Fe(II)	Fe(III)
0.00%-Mn	48.7%	51.3%
14.29%-Mn	57.8%	42.2%
25.00%-Mn	55.1%	44.9%

Table S5 Sb(III) and Sb(V) contents of Fe-Mn flocs at different Mn mass ratios

No.	Sb(III)	Sb(V)	Sb(III)/Sb(V)
0%-Mn	31.9%	68.1%	0.47
14.29%-Mn	36.7%	63.3%	0.58
25.00%-Mn	35.0%	65.0%	0.54

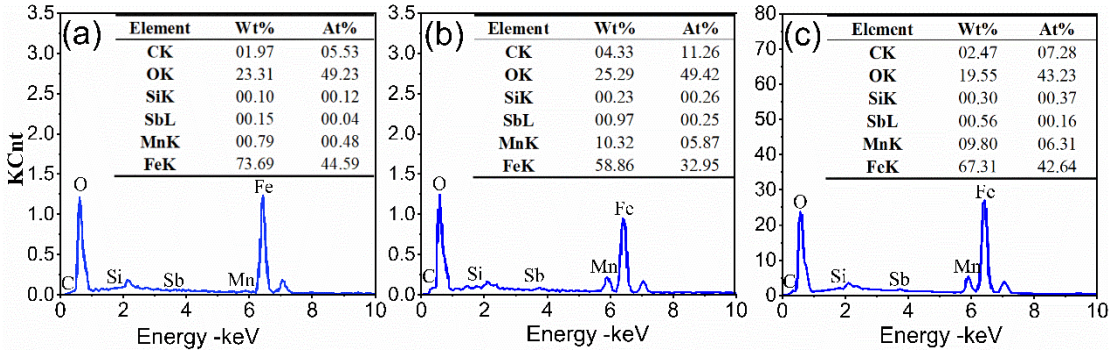

Fig. S1 EDS images of flocs (a) 0%-Mn (b) 14.29%-Mn (c) 25.00%-Mn

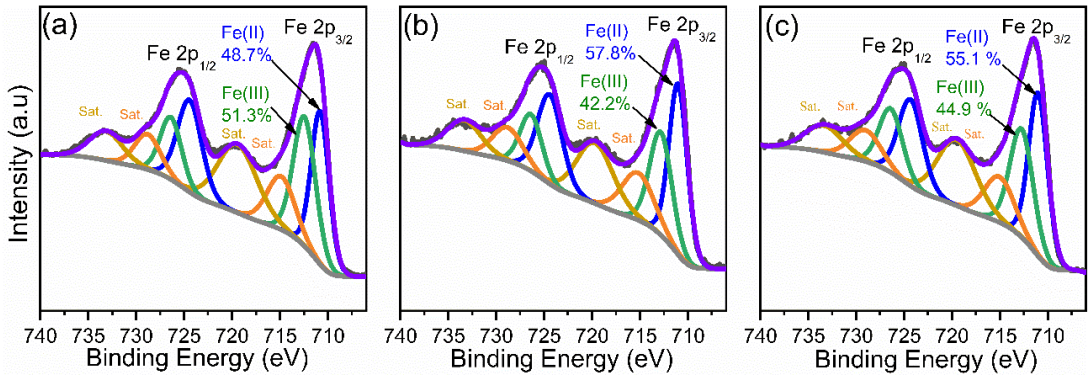

Fig. S2 High-resolution XPS spectra of Fe 2p of flocs (a) 0%-Mn (b) 14.29%-Mn (c) 25.00%-Mn

S1 Reaction equations involved in the Fe-Mn-C micro-electrolysis system

In the micro-electrolysis system, the presence of varying electrode potentials between iron and carbon, as well as manganese and carbon, facilitates redox reactions among the three distinct media (Eqs. (S1)-(S5)).

Anode (Fe & Mn):

$$Fe\text{-}2e^- \rightarrow Fe^{2+},\ E_0(Fe^{2+}/Fe)=-0.44V \quad (S1)$$

$$Fe^{2+}\text{-}e^- \rightarrow Fe^{3+},\ E_0(Fe^{3+}/Fe^{2+})=0.77V \quad (S2)$$

$$Mn\text{-}2e^- \rightarrow Mn^{2+},\ E_0(Mn^{2+}/Mn)=-1.88V \quad (S3)$$

Cathode (C):

$$2H^+ + 2e^- \rightarrow 2[H] \rightarrow H_2,\ E_0(H_2/H^+)= 0V \quad (S4)$$

$$O_2+2H_2O+4e^- \rightarrow 4OH^- \text{ (Alkaline or neutral conditions)},\ E_0(O_2/OH^-)=0.40\ V \quad (S5)$$

Iron and manganese also exhibit a discernible potential difference, rendering them susceptible to reaction (Eq. (S6)). As indicated by Table S4 and Fig. S1, the escalation in manganese content within the micro-electrolysis reaction system coincides with an elevation in the Fe^{2+} content within the flocs, concomitant with a reduction in Fe^{3+} concentration. This outcome signifies the occurrence of a supplementary redox reaction between metallic Mn and Fe^{3+} (Eq. (S7)).

$$Mn+Fe^{2+} \rightarrow Mn^{2+}+Fe \quad (S6)$$

$$Mn+2Fe^{3+} \rightarrow Mn^{2+}+2Fe^{2+} \quad (S7)$$

In the Sb(V) removal process, a portion of Sb(V) is adsorbed and eliminated by the iron-manganese flocs generated through micro-electrolysis, while another fraction undergoes reduction by Fe^{2+} and reductive hydrogen, resulting in the formation of $Sb(OH)_3$ precipitate during coagulation. This precipitate is subsequently removed through adsorption and co-precipitation reactions within the flocs (Eqs. (S8)-(S11)).

$$Mn^{2+}+FeOOH \rightarrow Mn_xFe_yOOH \quad (S8)$$

$$Sb^{5+}+2Fe^{2+} \rightarrow Sb^{3+}+2Fe^{3+} \quad (S9)$$

$$Sb^{5+}+2[H] \rightarrow Sb^{3+}+H_2 \quad (S10)$$

$$Sb^{3+}+OH^- \rightarrow Sb(OH)_3 \quad (S11)$$