

The form of Fe in solution and mineral

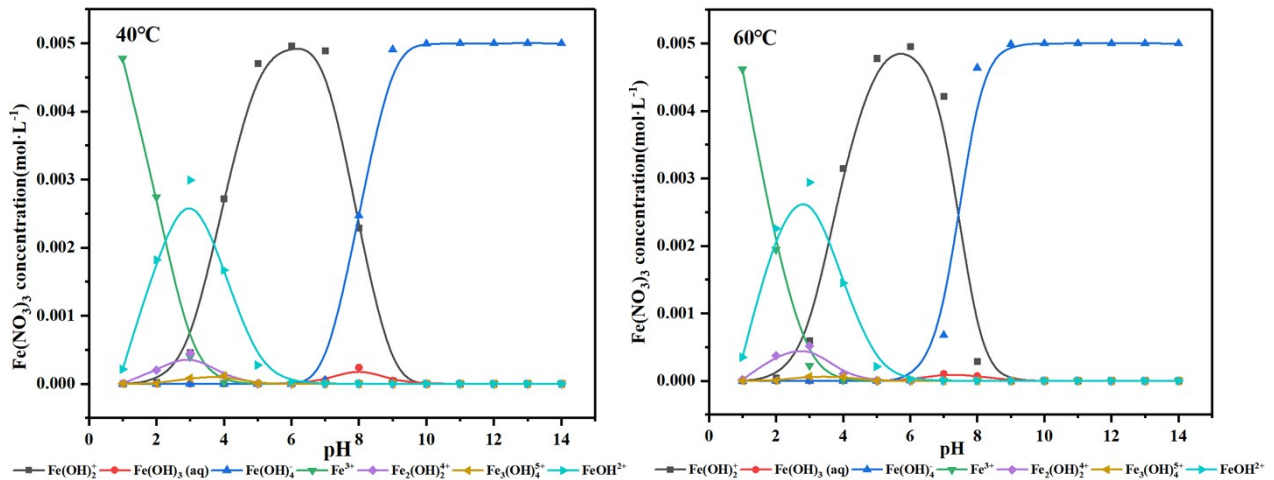


Fig. 1s Calculation results of the Fe(III) species distribution under variable pH conditions. Experiment conditions: concentration: $0.005\text{ mol}\cdot\text{L}^{-1}$ $\text{Fe}(\text{NO}_3)_3$, temperature: 40°C on the left, 60°C on the right, atmospheric pressure: 101.325 KPa .

Basic property of Pb-contaminated soils

Table 1s The Pb content and pH of the contaminated soils

Contaminated soil no.	Total Pb (mg/kg)	pH
1	4319.3	8.5
2	2544.6	7.13

Experimental conditions

Table 2s The detailed protocol for Pb washing experiments in mineral and soils

Group	Additive variety	Factor				
		Concentration /mol	Liquid-solid ratio	Temperature /°C	Time /h	
1	Inorganic acid agent (HCl)	0.01, 0.05, 0.1, 0.2, 0.4, 0.8	20	25	12	
	Washing agents (Na ₂ EDTA)	0.01, 0.05, 0.1, 0.2, 0.4	20	25	12	
	organic acid agent (citric acid)	0.01, 0.05, 0.1, 0.2, 0.4, 0.8	20	25	12	
2	Reductive agent selection	NH ₂ OH · HCl + HCl	NH ₂ OH · HCl: 0, 0.04, 0.1 HCl: 0, 0.4, 1	20	60	12
		VC + HCl	VC: 0, 0.05, 0.1 HCl: 0, 0.4, 1	20	60	12
				20	60	12

3	Mixed solution	VC + HCl	VC: 0, 0.01, 0.02, 0.05, 0.08, 0.1, 0.2 HCl: 0.01, 0.05, 0.1, 0.2, 0.4, 0.8	200	60	12
		VC + Na ₂ EDTA	VC: 0, 0.01, 0.02, 0.05, 0.08, 0.1, 0.2 Na ₂ EDTA: 0.01, 0.03, 0.05, 0.08, 0.1, 0.2	200	60	12
		VC + CA	VC: 0, 0.01, 0.02, 0.05, 0.08, 0.1, 0.2 CA: 0.01, 0.05, 0.1, 0.2, 0.4, 0.8	200	60	12
4	Liquid-solid ratio selection	VC + HCl	VC: 0.1 HCl: 0.1	10, 25, 50, 100, 150, 200	60	12
		VC + Na ₂ EDTA	VC: 0.05 Na ₂ EDTA: 0.05	10, 25, 50, 100, 150, 200	60	12
		VC + CA	VC: 0.08 CA: 0.1	10, 25, 50, 100, 150, 200	60	12
5	Temperature kinetics	VC + HCl	VC: 0.1 HCl: 0.1	150	25, 40, 60	5 min, 10 min, 20 min,
		VC + Na ₂ EDTA	VC: 0.05 Na ₂ EDTA: 0.05	200	25, 40, 60	40 min, 1h, 1.5h, 2 h, 4 h,
		VC + CA	VC: 0.08 CA: 0.1	150	25, 40, 60	6 h, 8 h and 12 h
6	Contaminated soils	VC + HCl	0.1 HCl, 0.1 HCl + 0.1VC, 0.4 HCl, 0.4 HCl + 0.05VC	150	60	12
		VC + Na ₂ EDTA	0.05 Na ₂ EDTA, 0.05 VC + 0.05Na ₂ EDTA	200	60	12
		VC + CA	0.1, 0.4	150	60	12
		VC	0.05, 0.08, 0.1	200	60	12

Table 3s The occurrence of Pb in the minerals (ug/g)

Pb in the Mineral	Exchangeable	Carbonate-bound	Fe-Mn oxides-bound	Organic-bound	Residual	Total content
Pb	0.8	463.09	20052.03	581.79	12046.33	33144.11

Washing experiment conditions and simulated results**Table 4s** The two reducing agents and concentrations on Pb removal efficiency

Test NO.	Mineral mass (g)	The volume of the mixed solution (ml)	HCl concentration	Reductant concentration	Temperature (°C)	Removal efficiency (%)	pH of the supernatant after washing
EX-1	0.1	20	0.4 mol·L ⁻¹ HCl	-	60	53.7	0.56
EX-2	0.1	20	-	0.04 mol·L ⁻¹ NH ₂ OH·HCl	60	23.6	3.6
EX-3	0.1	20	0.4 mol·L ⁻¹ HCl	0.04 mol·L ⁻¹ NH ₂ OH·HCl	60	61.2	0.56
EX-4	0.1	20	1 mol·L ⁻¹ HCl	0.04 mol·L ⁻¹ NH ₂ OH·HCl	60	88.8	0.18
EX-5	0.1	20	0.4 mol·L ⁻¹ HCl	0.1 mol·L ⁻¹ NH ₂ OH·HCl	60	61.4	0.55
EX-6	0.1	20	-	0.05 mol·L ⁻¹ VC	60	40.5	3.77
EX-7	0.1	20	0.4 mol·L ⁻¹ HCl	0.05 mol·L ⁻¹ VC	60	90.3	0.62
EX-8	0.1	20	1 mol·L ⁻¹ HCl	0.05 mol·L ⁻¹ VC	60	98.6	0.25
EX-9	0.1	20	0.4 mol·L ⁻¹ HCl	0.1 mol·L ⁻¹ VC	60	95.7	0.62

Table 5s Parameters of kinetics of Pb removal efficiency from iron oxide mineral to washing agents.

Washing agent	Temperature (°C)	Pseudo-first-order(x-1)			Pseudo-second-order(x-2)		
		Q ₁ (%)	k ₁ (% ⁻¹ h ⁻¹)	R ₁ ²	Q ₂ (%)	k ₂ (% ⁻¹ h ⁻¹)	R ₂ ²
a (0.1M HCl+0.1M VC)	25	42.78	1.1521	0.991*	47.57	0.0321	0.977*
	40	76.85	2.5112	0.980*	81.92	0.0477	0.949*
	60	98.76	3.9374	0.912*	104.34	0.0598	0.969*
b (0.05M Na ₂ EDTA+0.05M VC)	25	45.52	2.1323	0.980*	49.01	0.0646	0.967*
	40	70.61	3.0720	0.982*	75.07	0.0630	0.969*
	60	98.59	4.3372	0.939*	103.55	0.0687	0.964*
c (0.08M CA+0.1M VC)	25	47.49	0.5176	0.958*	58.05	0.0090	0.935*
	40	65.51	3.1804	0.942*	69.88	0.0683	0.975*
	60	95.21	2.3004	0.805*	101.62	0.0371	0.925*

*Significant at P=0.01 level

Table 6s Experimental conditions of contaminated soils

Group no.	Acid washing agents (mol·L ⁻¹)	VC (mol·L ⁻¹)	Time (h)
1-untreated	-	-	-
2	0.1mol·L ⁻¹ CA	-	12
3	0.1mol·L ⁻¹ CA	0.08	12
4	0.4mol·L ⁻¹ CA	-	12
5	0.4mol·L ⁻¹ CA	0.05	12
6	0.1mol·L ⁻¹ HCl	-	12
7	0.1mol·L ⁻¹ HCl	0.1	12
8	0.4mol·L ⁻¹ HCl	-	12
9	0.4mol·L ⁻¹ HCl	0.05	12
10	0.05mol·L ⁻¹ Na ₂ EDTA	-	12
11	0.05mol·L ⁻¹ Na ₂ EDTA	0.05	12
12	-	0.05	12
13	-	0.08	12
14	-	0.1	12