

Supplementary Information

Process Design and Validation of A New Mixed Eluent for Leaching Cd, Cr, Pb, Cu, Ni and Zn from Heavy Metal Polluted Soil

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Table S1 Operating parameters of ICP-MS and FTIRS

Parameter	Setting
ICP-MS	
ICP power	1150 W
Sampling cone aperture	1.2 mm
Cooling air flow	18 L/min
Intercepting cone aperture	1.0 mm
Auxiliary gas flow rate	1.2 L/min
Number of scans	20 times
Atomization gas flow rate	0.92 L/min
Measuring time	60 S
FTIRS	
wave-number range	4000 ~ 400 cm^{-1}
spectral resolution	0.4 cm^{-1}
wave number precision	0.01 cm^{-1}

Table S2 Heating program of microwave digestion

Step	Heating-up time (min)	Target temperature (°C)	Retention time (min)	Pressure(MPa)
1	5	120	2	1.5
2	4	150	5	2.0
3	5	185	40	2.0

Table S3 Summary for the measurement results of Cd, Cr, Cu, Ni, Pb and Zn in standard solutions

Element	Concentration ($\mu\text{g/mL}$)	Linear range ($\mu\text{g/mL}$)	Linear equation	R
Cd	0.000, 0.500, 2.000, 5.000, 20.000	0.000-20.000	$y=69.022x$	0.9999
Cr	0.000, 0.500, 2.000, 5.000, 20.000	0.000-20.000	$y=351.230x+3410.22$	0.9998
Pb	0.000, 0.500, 2.000, 5.000, 20.000	0.000-20.000	$y=358.274x$	0.9997
Cu	0.000, 0.500, 1.000, 2.000, 5.000	0.000-5.000	$y=141.813x$	0.9999
Zn	0.000, 0.500, 1.000, 2.000, 5.000	0.000-5.000	$y=256.598x$	0.9998
Ni	0.000, 0.500, 1.000, 2.000, 5.000	0.000-5.000	$y=69.022x$	0.9999

R: correlation coefficient.

Table S4 Detection limit of ICP-MS method

Element	Cd ($\mu\text{g/g}$)	Cr ($\mu\text{g/g}$)	Pb ($\mu\text{g/g}$)	Cu ($\mu\text{g/g}$)	Ni ($\mu\text{g/g}$)	Zn ($\mu\text{g/g}$)
C1	0.0013	0.0221	0.0739	0.0208	0.0035	0.0272
C2	0.0022	0.0214	0.0756	0.0147	0.0031	0.0274
C3	0.0013	0.0221	0.0765	0.0344	0.0033	0.0225
C4	0.0007	0.0196	0.0910	0.0287	0.0064	0.0258
C5	0.0035	0.0151	0.0893	0.0249	0.0029	0.0168
C6	0.0158	0.0232	0.0813	0.0313	0.0041	0.0209
C7	0.0017	0.0207	0.0588	0.0184	0.0058	0.0292
C8	0.0029	0.0243	0.0611	0.0432	0.0058	0.0024
C9	0.0011	0.0215	0.0392	0.0375	0.0044	0.0237
C10	0.0012	0.0219	0.0809	0.0481	0.0083	0.0336
C11	0.0021	0.0239	0.0304	0.0304	0.0036	0.0166
C12	0.0287	0.0247	0.0686	0.0338	0.0094	0.0332
Average	0.0052	0.0217	0.0689	0.0305	0.0051	0.0233
SD	0.0085	0.0026	0.0187	0.0099	0.0021	0.0087
DL	0.0254	0.0077	0.0562	0.0296	0.0064	0.0257

SD: standard deviation of the blank; DL: detection limit, which is estimated as three times standard deviation of the blank.

Table S5 Determination of soil pH

Soil sample number	1	2	3	4	5	6	7	8	9	10
pH	8.36	8.35	8.35	8.36	8.36	8.37	8.36	8.37	8.36	8.36
Average					8.36					
SD					0.01					
RSD (%)					0.08					

Table S6 Leaching rates of heavy metals by different chelating agents

Eluent	Cd		Cr		Pb		Cu		Ni		Zn	
	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)
0.050 mol/L EDDS	0.86 \pm 0.05	19.39	2.08 \pm 0.02	6.23	409.31 \pm 0.32	23.34	53.00 \pm 0.32	26.90	0.06 \pm 0.10	0.30	93.70 \pm 0.10	11.71
0.040 mol/L EDTA	0.88 \pm 0.18	19.80	2.24 \pm 0.74	6.73	371.31 \pm 0.81	21.24	47.50 \pm 0.83	24.11	0.48 \pm 0.94	2.40	95.74 \pm 0.30	11.96
0.005 mol/L DTPA	0.98 \pm 0.03	22.11	1.23 \pm 0.17	3.70	442.80 \pm 0.91	25.31	51.51 \pm 0.56	26.14	0.90 \pm 0.76	4.55	97.20 \pm 0.13	12.15
0.050 mol/L GLDA	0.05 \pm 0.02	1.05	0.00 \pm 0.00	0.00	14.30 \pm 0.09	0.88	25.03 \pm 0.32	12.69	0.05 \pm 0.01	0.25	18.24 \pm 0.01	2.27
0.100 mol/L PASP	0.31 \pm 0.05	7.07	0.43 \pm 0.02	1.29	38.34 \pm 0.40	2.25	38.50 \pm 0.80	19.54	0.30 \pm 0.57	1.52	49.67 \pm 0.02	6.20
0.060 mol/L SDBS	0.00 \pm 0.00	0.00	0.17 \pm 0.12	0.52	2.43 \pm 0.21	0.20	4.50 \pm 0.29	1.79	0.01 \pm 0.01	0.05	6.77 \pm 0.03	0.84
0.010 mol/L IDS	0.30 \pm 0.01	6.73	0.01 \pm 0.02	0.03	127.3 \pm 0.40	7.33	37.52 \pm 0.23	19.03	1.60 \pm 0.31	8.08	58.73 \pm 0.20	7.34
0.010 mol/L STPP	1.04 \pm 0.04	23.32	3.88 \pm 0.14	11.64	234.31 \pm 0.37	13.43	16.51 \pm 0.82	8.37	0.00 \pm 0.00	0.00	87.75 \pm 0.01	10.96

Note: The concentration of each chelating agent is the best content selected from previous reports. The solution pH value is 6.50, the liquid-solid ratio is 10:1, the oscillation rate is 100 rpm, the oscillation time is 12 h, and the centrifugation time is 40 min.

Table S7 Leaching efficiency of EDDS for heavy metals under different concentrations

Concentration (mol/L)	Cd		Cr		Pb		Cu		Ni		Zn	
	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)
0.010	0.54 ± 0.04	12.19	0.14 ± 0.03	0.42	89.34 ± 0.98	5.10	30.46 ± 0.13	15.46	0.94 ± 0.17	4.75	22.27 ± 0.04	2.85
0.030	0.91 ± 0.01	20.52	2.10 ± 0.02	6.29	449.87 ± 0.48	25.65	40.81 ± 0.63	21.22	1.71 ± 0.03	8.65	35.97 ± 0.56	4.51
0.050	0.86 ± 0.05	19.39	2.08 ± 0.02	6.23	409.31 ± 0.32	23.34	53.00 ± 0.32	26.90	0.06 ± 0.10	0.30	93.70 ± 0.10	11.71
0.060	0.91 ± 0.03	20.41	2.09 ± 0.58	6.26	417.47 ± 0.40	23.81	31.63 ± 0.47	16.06	1.12 ± 0.11	5.63	24.30 ± 0.30	3.04
0.070	1.24 ± 0.06	27.89	4.90 ± 0.05	14.70	519.42 ± 0.25	29.62	38.55 ± 0.32	19.56	1.95 ± 0.23	9.85	43.87 ± 0.05	5.49
0.080	0.81 ± 0.03	18.29	2.58 ± 0.21	7.76	350.94 ± 0.85	20.01	31.22 ± 0.23	15.82	1.10 ± 0.06	5.55	24.71 ± 0.50	3.09
0.090	0.86 ± 0.01	19.46	3.27 ± 0.48	9.83	416.53 ± 0.18	23.75	34.92 ± 0.18	17.72	1.38 ± 0.16	6.96	28.43 ± 0.09	3.56
0.100	0.98 ± 0.08	22.14	3.84 ± 0.22	11.52	464.85 ± 0.53	26.51	41.70 ± 0.29	21.17	1.82 ± 0.10	9.17	33.34 ± 0.40	4.17

Note: The liquid-solid ratio was 10:1, the oscillation rate was 100 rpm, the oscillation time was 12 h, and the centrifugation time was 40 min. The pH is 6.50.

Table S8 Leaching effect of heavy metal elements by FeCl₃

Eluent	Content(mol/L)	Cd		Pb	
		Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)
FeCl ₃	0.010	0.29 \pm 4.00	6.59	64.90 \pm 2.44	3.70
	0.050	0.76 \pm 2.28	17.05	444.46 \pm 0.23	25.34
	0.100	3.21 \pm 0.62	68.05	666.70 \pm 1.43	38.01
	0.150	4.13 \pm 1.29	93.05	629.69 \pm 0.74	35.90
	0.200	0.73 \pm 1.83	16.48	652.84 \pm 0.96	37.22

Note: The liquid-solid ratio was 10:1, the oscillation rate was 100 rpm, the oscillation time was 12 h, and the centrifugation time was 40 min in the experiment. The pH is the original pH.

Table S9 Leaching efficiency of EDDS for heavy metals under different pH conditions

pH	Cd		Cr		Pb		Cu		Ni		Zn	
	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)	Content ($\mu\text{g/g}$)	Efficiency (%)
1.50	1.24 \pm	28.03	3.18 \pm	9.55	222.67 \pm	12.69	33.11 \pm	16.81	1.77 \pm	8.94	31.86 \pm	3.98
	0.15		0.49		0.94		0.51		0.36		0.77	
2.50	0.87 \pm	19.59	2.22 \pm	6.67	66.19 \pm	3.77	29.97 \pm	15.21	1.57 \pm	7.91	25.23 \pm	3.15
	0.01		0.12		0.04		0.07		0.03		0.31	
3.50	0.78 \pm	17.62	2.52 \pm	7.58	70.68 \pm	4.03	33.67 \pm	17.09	1.68 \pm	8.50	27.56 \pm	3.45
	0.19		0.47		0.51		0.36		0.26		0.22	
4.50	0.72 \pm	16.11	2.60 \pm	7.82	306.59 \pm	17.48	26.73 \pm	13.57	1.24 \pm	6.27	22.39 \pm	2.80
	0.14		0.32		0.50		1.00		0.18		0.78	
5.50	0.97 \pm	21.88	2.94 \pm	8.82	413.54 \pm	23.58	32.31 \pm	16.40	1.41 \pm	7.11	24.16 \pm	3.02
	0.19		0.17		1.61		0.26		0.37		0.43	
6.50	0.86 \pm	19.39	2.08 \pm	6.23	409.31 \pm	23.34	53.00 \pm	26.90	0.06 \pm	0.30	93.70 \pm	11.71
	0.05		0.02		0.32		0.32		0.10		0.10	
7.50	0.92 \pm	20.78	2.49 \pm	7.47	385.56 \pm	21.98	32.36 \pm	16.43	1.36 \pm	6.89	22.64 \pm	2.83
	0.20		0.25		0.56		0.58		0.02		0.10	
8.50	0.84 \pm	18.99	2.57 \pm	7.71	382.41 \pm	21.80	33.18 \pm	16.84	1.33 \pm	6.69	21.92 \pm	2.74
	0.07		0.12		0.88		0.15		0.09		1.08	
9.50	0.90 \pm	20.32	2.63 \pm	7.91	391.40 \pm	22.31	35.27 \pm	17.90	1.41 \pm	7.11	22.49 \pm	2.81
	0.08		0.01		0.74		0.66		0.07		0.44	
10.50	0.94 \pm	21.23	2.73 \pm	8.19	399.85 \pm	22.80	36.40 \pm	18.48	1.46 \pm	7.36	22.22 \pm	2.78
	0.11		0.08		0.06		0.58		2.23		0.07	

Note: The concentration of EDDS was 0.050 mol/L, the liquid-solid ratio was 10:1, the oscillation rate was 100 rpm, the oscillation time was 12 h, and the centrifugation time was 40 min.

Table S10 Leaching efficiency of the mixed eluent EDDS and FeCl₃

m _{EDDS} : m _{FeCl₃}	Cd		Cr		Pb		Cu		Ni		Zn		pH	
	Content (µg/g)	Efficiency (%)	Content (µg/g)	Efficiency (%)	Content (µg/g)	Efficiency (%)	Content (µg/g)	Efficiency (%)	Content (µg/g)	Efficiency (%)	Content (µg/g)	Efficiency (%)		
1:5	2.04±	46.25	3.56±	10.72	411.14±	23.44	32.11±	16.27	0.44±	2.23	8.73±	1.09	Original	
	0.52		0.29		0.39		0.37		0.13		0.11			
3:5	2.54±	57.64	3.14±	9.43	243.11±	13.86	22.99±	11.65	0.14±	0.69	15.69±	1.96		
	0.42		0.13		0.21		0.16		0.20		0.13			
1:1	2.12±	47.79	6.01±	18.03	460.61±	26.26	50.78±	25.73	4.85±	24.55	22.82±	2.85		pH
	0.42		0.35		0.25		0.22		0.25		0.19			
9:5	02.41±	54.88	4.79±	14.35	435.17±	24.81	31.68±	16.05	0.23±	1.14	16.65±	2.08		
	0.47		0.26		0.40		0.27		0.19		0.29			
21:5	2.83±	64.29	3.59±	10.75	404.30±	23.05	50.23±	25.45	3.36±	16.95	33.07±	4.13		
	0.51		0.22		0.32		0.21		0.16		0.06			
1:5	3.17±	71.36	4.08±	12.25	431.75±	24.62	36.93±	18.74	0.41±	2.07	13.44±	1.68	1.30	
	0.64		0.36		0.42		0.31		0.28		0.11			
3:5	3.10±	69.80	3.43±	10.29	398.52±	22.72	28.22±	14.32	0.24±	1.20	16.19±	2.02		
	0.47		0.26		0.37		0.29		0.07		0.15			
1:1	3.02±	67.97	7.09±	21.29	513.93±	29.30	56.87±	28.87	6.74±	34.05	35.21±	4.40		
	0.62		0.30		0.35		0.35		0.47		0.14			
9:5	3.16±	71.25	6.12±	18.38	550.92±	31.41	32.93±	16.72	0.27±	1.35	20.42±	2.55		
	0.64		0.32		0.23		0.12		0.14		0.19			
21:5	3.11±	69.97	4.56±	13.71	506.26±	28.86	50.48±	25.63	3.53±	17.80	36.24±	4.53		
	0.64		0.30		0.36		0.35		0.23		0.19			

1:5	1.56± 0.58	35.13	0.02± 0.03	0.06	80.33± 0.25	4.58	52.39± 0.45	26.59	4.49± 0.43	22.66	35.95± 0.17	4.49	
3:5	2.11± 0.44	47.61	3.43± 0.01	0.19	113.82± 0.17	6.49	49.76± 0.24	25.26	3.43± 0.20	17.32	33.23± 0.15	4.15	
1:1	1.21± 0.43	27.15	0.07± 0.23	0.22	79.05± 0.22	4.51	33.96± 0.31	17.24	3.07± 0.27	15.49	15.58± 0.52	1.95	6.50
9:5	1.89± 0.55	42.65	0.38± 0.11	1.15	129.92± 0.23	7.41	59.60± 0.53	30.25	5.74± 0.24	29.00	39.66± 0.19	4.96	
21:5	2.56± 0.52	57.72	0.27± 0.13	0.81	313.15± 0.27	17.85	46.92± 0.38	23.81	2.48± 0.21	12.52	23.21± 0.06	2.90	
1:5	0.04± 0.06	0.97	0.23± 0.09	0.69	127.67± 0.15	7.28	35.05± 0.31	17.77	2.18± 0.19	11.00	17.54± 0.23	2.19	
3:5	0.06± 0.19	1.33	0.67± 0.27	2.01	359.70± 0.37	20.51	38.93± 0.19	19.76	1.90± 0.26	9.61	21.57± 0.30	2.70	
1:1	1.94± 0.50	43.72	0.25± 0.12	0.74	191.61± 0.30	10.92	46.92± 0.36	23.82	3.63± 0.15	18.34	17.01± 0.16	2.13	8.50
9:5	0.74± 0.34	16.70	0.72± 0.35	2.17	498.40± 0.37	28.42	42.98± 0.45	21.82	2.96± 0.23	14.96	19.98± 0.13	2.50	
21:5	2.38± 0.50	53.60	0.35± 0.06	1.04	328.03± 0.29	18.70	46.33± 0.37	23.52	2.10± 0.23	10.62	23.26± 0.16	2.91	

Note: $m_{\text{EDDS}}: m_{\text{FeCl}_3}$ is the mass ratio of EDDS and FeCl_3 . The concentration of EDDS was 0.050 mol/L, the concentration of FeCl_3 was 0.150 mol/L, the liquid-solid ratio was 10:1, the oscillation rate was 100 rpm, the oscillation time was 12 h, and the centrifugation time was 40 min.

Table S11 Physical and chemical properties of soil before and after leaching.

Characteristics	Original soil	Soil after EDDS+ FeCl ₃ leaching															Soil after FeCl ₃ leaching
		pH=1.30					pH=6.50					pH=8.50					
		1:5	3:5	1:1	9:5	21:5	1:5	3:5	1:1	9:5	21:5	1:5	3:5	1:1	9:5	21:5	
pH	8.36± 0.01	4.32± 0.01	4.35± 0.01	4.56± 0.01	4.74± 0.01	4.94± 0.01	5.51± 0.01	5.63± 0.01	6.25± 0.01	6.67± 0.01	6.79± 0.01	8.38± 0.01	8.64± 0.01	8.89± 0.01	8.94± 0.01	8.91± 0.01	3.37 ± 0.01
SOM (%)	5.02± 0.04	2.24± 0.01	2.26± 0.01	2.28± 0.02	2.25± 0.01	2.27± 0.02	2.45± 0.02	2.66± 0.02	2.64± 0.01	2.81± 0.02	3.02± 0.04	2.43± 0.02	2.70± 0.02	2.67± 0.02	2.89± 0.02	2.54± 0.02	2.25 ± 0.16
N (mg/g)	3.47± 0.35	2.34± 0.15	2.38± 0.16	2.35± 0.15	2.46± 0.15	2.44± 0.15	2.52± 0.17	2.57± 0.15	2.49± 0.15	2.55± 0.16	2.63± 0.15	2.68± 0.18	2.72± 0.15	2.79± 0.19	2.90± 0.21	2.84± 0.15	2.02 ± 0.08
P (µg/g)	54.40± 0.03	25.18± 0.01	25.23± 0.01	25.19± 0.01	25.64± 0.01	35.88± 0.02	27.07± 0.01	36.59± 0.03	29.48± 0.01	31.60± 0.01	38.06± 0.02	34.38± 0.01	28.94± 0.01	48.38± 0.03	40.25± 0.02	46.32± 0.01	22.14 ± 0.02

SOM: soil organic matter; N: Nitrogen; P: Phosphorus.