Electronic Supplementary Information

Impurity removal with highly selective and efficient methods and recycling of transition metals from spent lithium-ion batteries

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Table S1 The list of metal ions' concentrations after adjusting the pH values.[a]

pH value	Aluminum (ppm)	Copper (ppm)	Iron (ppm)	Manganese (ppm)	Cobalt (ppm)	Nickel (ppm)
Original solution	613.5	697.9	587.9	5426	3234	6376
2.6	613.8	698.2	340	5414	3239	6368
2.9	612.6	697.6	287.2	5419	3224	6383
3.2	612.8	696.7	12.8	5421	3227	6385
3.3	614.3	698.8	6	5406	3218	6376
3.4	612.7	696.8	1.2	5424	3240	6372
3.5	612.9	698.3	0.2	5419	3242	6373
3.6	612.5	697.2	＜0.2	5416	3228	6368
3.7	612.6	698.1	＜0.2	5418	3223	6369
4.90	86.4	616.9	＜0.2	5413	3228	6334
5.00	59.7	611.8	＜0.2	5351	3186	6251
5.10	28.6	607.1	＜0.2	5371	3197	6300
5.20	3.8	604.5	＜0.2	5444	3241	6375
5.25	1.4	586	＜0.2	5413	3242	6372
5.30	1.5	588.3	＜0.2	5431	3213	6344
5.35	2.3	585.8	＜0.2	5434	3240	6389
5.40	1.9	578.6	＜0.2	5412	3238	6375
5.45	1.7	572.9	＜0.2	5395	3236	6353
5.50	1.6	559.3	＜0.2	5444	3236	6399
5.55	0.5	538	＜0.2	5379	3242	6348
5.65	＜0.3	474.4	＜0.2	5428	3234	6318

5.75	＜0.3	449.6	＜0.2	5405	3202	6293
5.85	＜0.3	434.5	＜0.2	5369	3208	6280
5.95	＜0.3	400.7	＜0.2	5358	3204	6268
6.05	＜0.3	256.5	＜0.2	5272	3165	6204
6.15	＜0.3	173	＜0.2	5374	3199	6239
6.25	＜0.3	127.8	＜0.2	5286	3188	6195
6.35	＜0.3	5.6	＜0.2	5309	3066	5903
6.45	＜0.3	＜0.04	＜0.2	5312	3015	5898
6.55	＜0.3	＜0.04	＜0.2	5320	2958	5888
6.65	＜0.3	＜0.04	＜0.2	5351	2920	5752
6.75	＜0.3	＜0.04	＜0.2	5252	2775	5552

[a] The pH values from 2.6 to 3.7 is the stage of removing Fe^{3+} ; the pH values from 4.9 to 5.75 is the stage of removing Al^{3+}; the pH values from 5.85 to 6.75 is the stage of removing Cu^{2+}.

Table S2 The possible reduction reactions in the metal solution and their standard electrode potentials (25 ℃).

Electrode reaction	Standard electrode potential (V vs. SHE)	Standard electrode potential (V vs. MSE)
$Mn^{2+}+2e^{-}=Mn$	-1.180	-1.796
$Co^{2+}+2e^{-}=Co$	-0.277	-0.893
$Ni^{2+}+2e^{-}=Ni$	-0.250	-0.866

$2H^{+}+2e^{-}=H_2$	0.000	-0.616
$Cu^{2+}+e^{-}=Cu^{+}$	0.153	-0.463
$Cu^{2+}+2e^{-}=Cu$	0.337	-0.279
$Cu^{+}+e^{-}=Cu$	0.521	-0.095

Table S3 Effect of different concentrations of extractant P on metal ions in the process of copper removal by solvent extraction.

concentration	Copper (ppm)	Manganese (ppm)	Cobalt (ppm)	Nickel (ppm)
Original solution	682.4	5336	3194	6306
5%	＜0.04	5339	3208	6318
15%	＜0.04	5332	3185	6213
30%	＜0.04	5326	3198	6298
45%	＜0.04	5344	3210	6294

Table S4 The concentrations of metal elements in the solution before and after electrodeposition.

Solution	Copper (ppm)	Manganese (ppm)	Cobalt (ppm)	Nickel (ppm)
Before electrodeposition	682.4	5336	3198	6306
After electrodeposition	6.1	5321	3211	6295