minerals

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Supplementary Material

Selective leaching of lithium and beyond: sustainable eggshell-mediated recovery from spent Li-ion batteries

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Table S1. Weight analysis on various parts of selected batteries.

Mass (g)						Weight percent (%)				
Battery	Anode	Cathode	Plastics	Steel	Connectors	An-ode	Cath-ode	Plas-tics	Steel	Connect-ors
37.93	10.13	23.99	0.89	1.76	0.3	26.71	63.25	2.35	4.64	0.79
34.8	18.15	12.8	1.46	1.95	0.44	52.16	36.78	4.20	5.60	1.26
16.5	5.08	7.64	2.022	0.93	0.23	30.79	46.30	12.25	5.64	1.39
53.66	18.32	28.4	0.54	2.03	1.33	34.14	52.93	1.01	3.78	2.48
23.94	6.53	9.38	2.58	5.11	0.3	27.28	39.18	10.78	21.35	1.25
30.8	10.2	13.03	2.25	5.3	0.06	33.12	42.31	7.31	17.21	0.19
24.42	6.54	8.67	2.66	4.46	0.18	26.78	35.50	10.89	18.26	0.74
27.81	6.22	11.57	2.15	5.3	0.33	22.37	41.60	7.73	19.06	1.19
21.3	5.67	7.3	3.02	4.02	0.3	26.62	34.27	14.18	18.87	1.41
15.56	4.41	5.27	1.17	2.69	0.3	28.34	33.87	7.52	17.29	1.93
27.27	7.27	10.08	2	4.69	0.3	26.66	36.96	7.33	17.20	1.10
41.39	11.65	16.82	4.07	6.9	0.5	28.15	40.64	9.83	16.67	1.21
14.5	3.02	4.07	2.48	4.28	0.3	20.83	28.07	17.10	29.52	2.07
19.13	7.16	10.65	2.02	2.24	0.3	37.43	55.67	10.56	11.71	1.57
18.17	2.93	4.68	2.15	2.91	0.2	16.13	25.76	11.83	16.02	1.10
26.96	6.44	12.24	2.21	3.83	0.2	23.89	45.40	8.20	14.21	0.74
Average (%)						28.84	41.16	8.94	14.81	1.28

Table S2. Summary of weight analyses for the entire collection of batteries.

	Anode	Cathode	Plastics	Steel	Connectors
Weight (g)	675.17	895.04	136.62	233.7	28.37
Weight percent (%)	30.35	40.23	6.14	10.50	1.28

Table S3. Optimal results of reductive acid leaching to achieve the highest Co leaching efficiency.

Co (%)	Mn (%)	Ni (%)	Li (%)
80.29	100	97.78	99.75

Table S4. pH changes over 1-minute intervals: equilibrium and adjusted values.

Time (min)	Equilibrium pH (after 1min intervals)	Adjusted pH after equilibrium
0	-	6.4
1	3.54	4.4
2	3.6	4.95
3	3.8	5.45
4	4.1	5.45
5	4.2	5.8
6	4.4	-

Table S5. Extraction efficiency with pH adjustment after 6 minutes.

Co (%)	Ni (%)	Li (%)
36.88	6.52	5.34

Table S6. Ni extraction and stripping results at different saponification degrees (Extraction conditions: O/A = 1, n-Heptane/CYANEX 272 = 4, room temperature, 10 minutes. Stripping conditions: O/A = 1, room temperature, and 5 minutes).

	Stage	Ni	Co	Li
Saponification degree of 10%	**Extraction (%)**	99.12	99.76	0.8
	Stripping (%)	95.18	100	100
Saponification degree of 20%	**Extraction (%)**	99.31	99.83	21.24
	Stripping (%)	85.85	98.86	100



Figure S1. A view of the collected spent Li-ion batteries from different mobile phones.

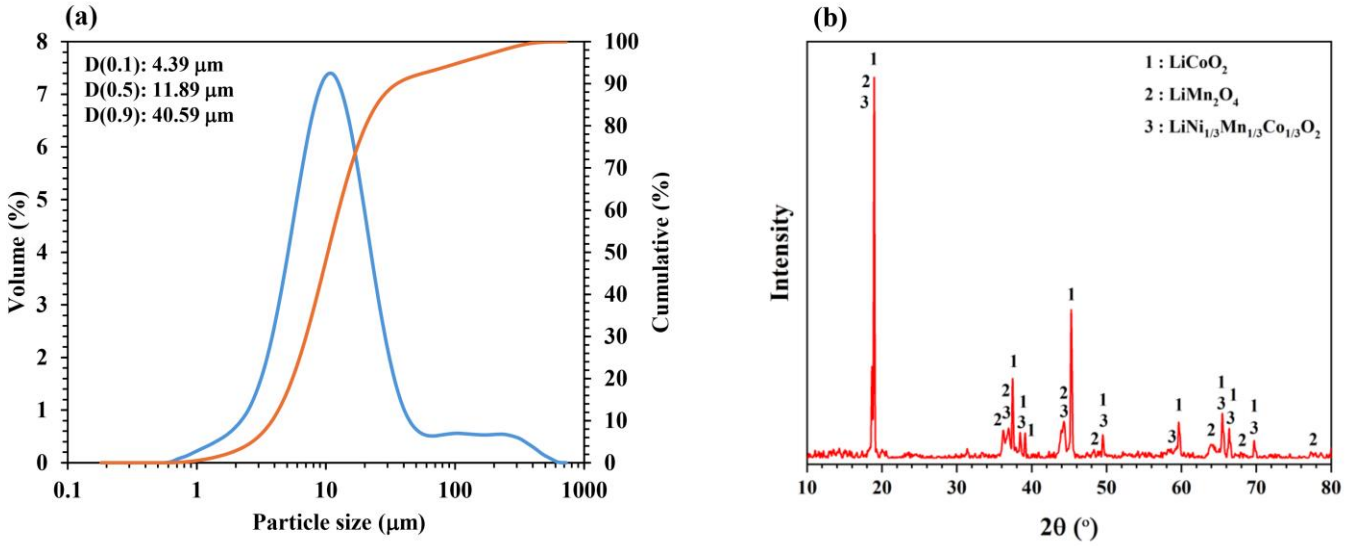


Figure S2. (**a**) Particle size distribution of the cathode active material and (**b**) XRD pattern depicting cathode active materials present in mixed batteries.

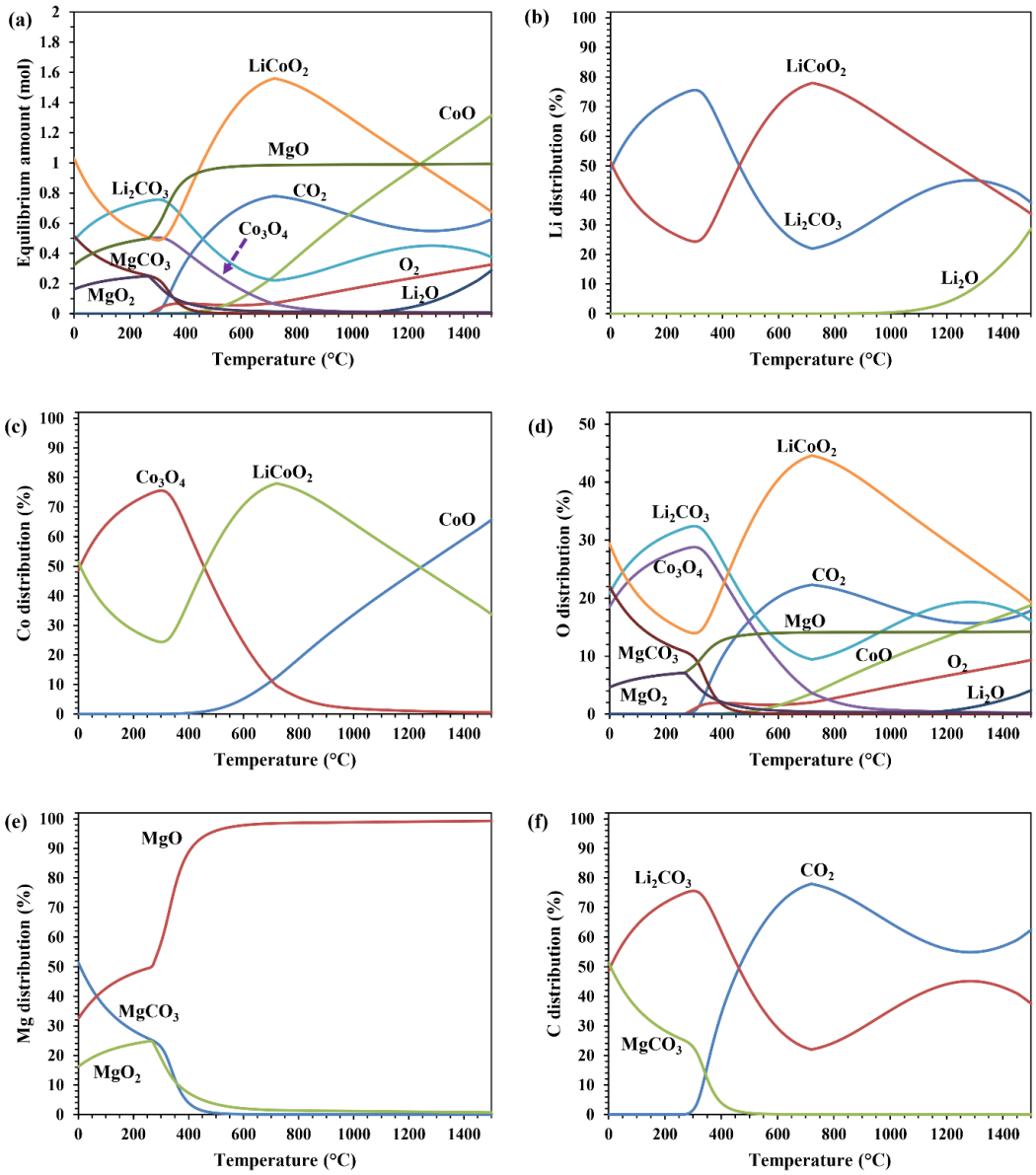


Figure S3. Equilibrium composition between 2 moles of $LiCoO_2$ and 1 mole of magnesium carbonate versus temperature predicted by HSC Chemistry® 10: (a) equilibrium amounts of possible products, (b) lithium distribution between different components, (c) cobalt distribution between different components, (d) oxygen distribution between different components, (e) magnesium distribution between different components, (f) carbon distribution between different components.

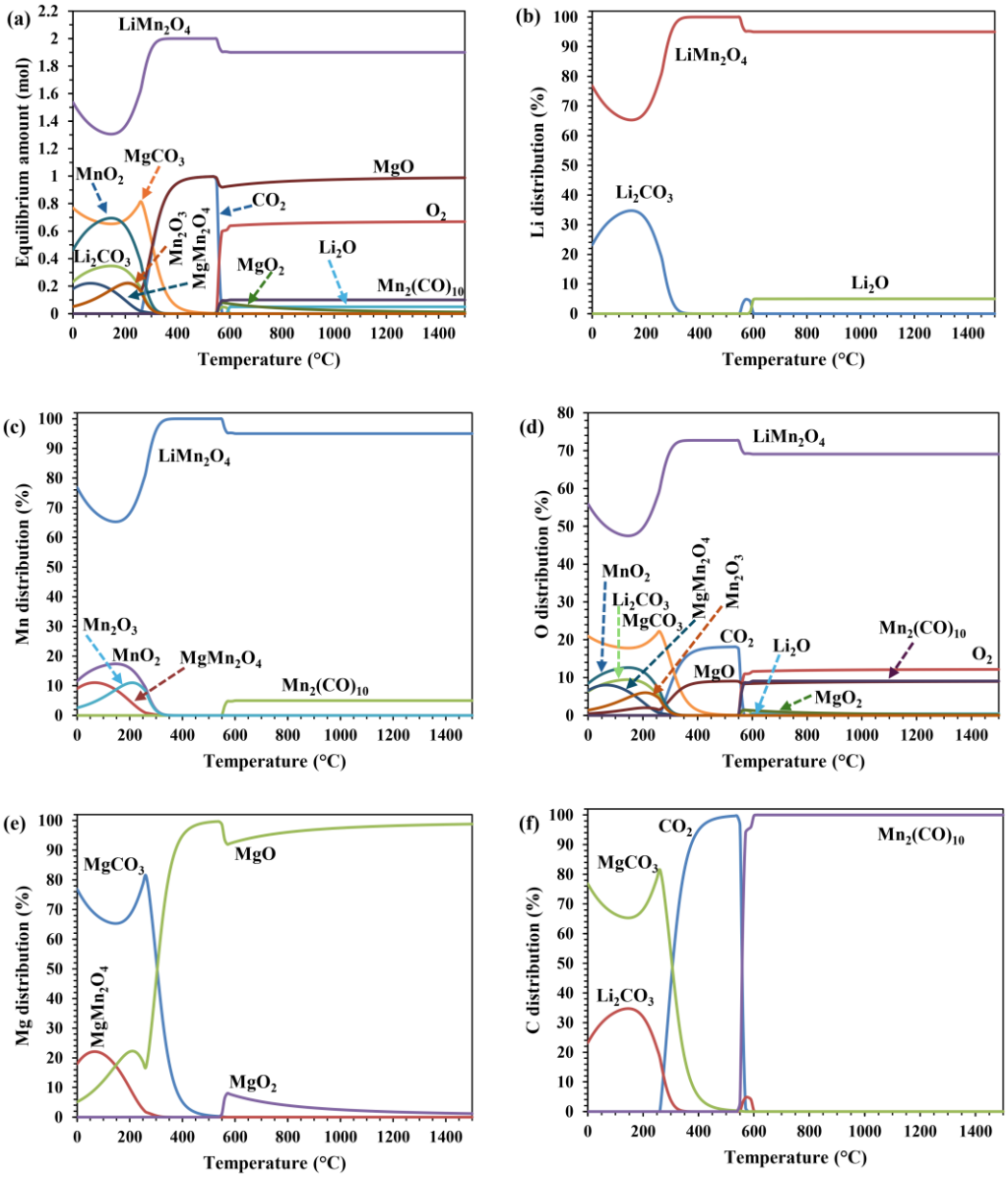


Figure S4. Equilibrium composition between 2 moles of $LiMn_2O_4$ and 1 mole of magnesium carbonate versus temperature predicted by HSC Chemistry® 10: (a) equilibrium amounts of possible products, (b) lithium distribution between different components, (c) manganese distribution between different components, (d) oxygen distribution between different components, (e) magnesium distribution between different components, (f) carbon distribution between different components.

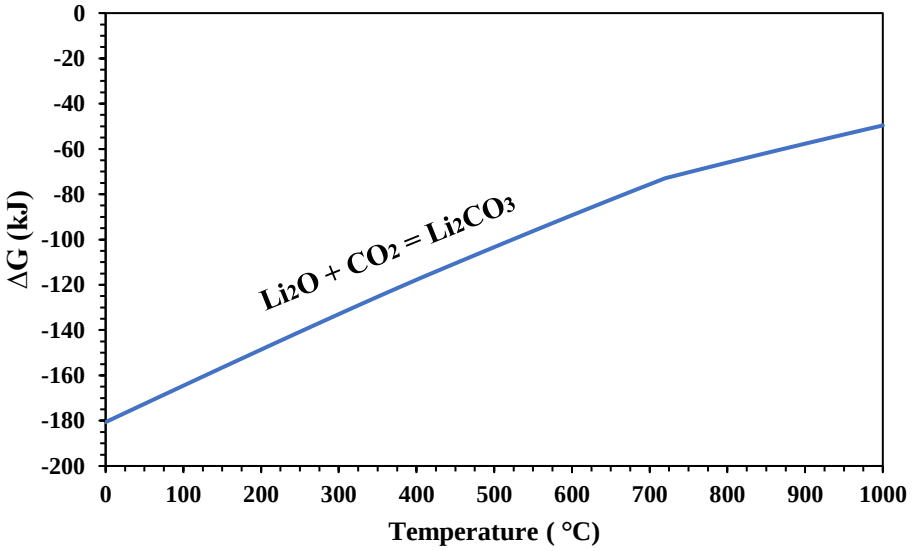


Figure S5. Variation of Gibbs free energy versus temperature for lithium carbonate formation in the absence of mechanical milling.

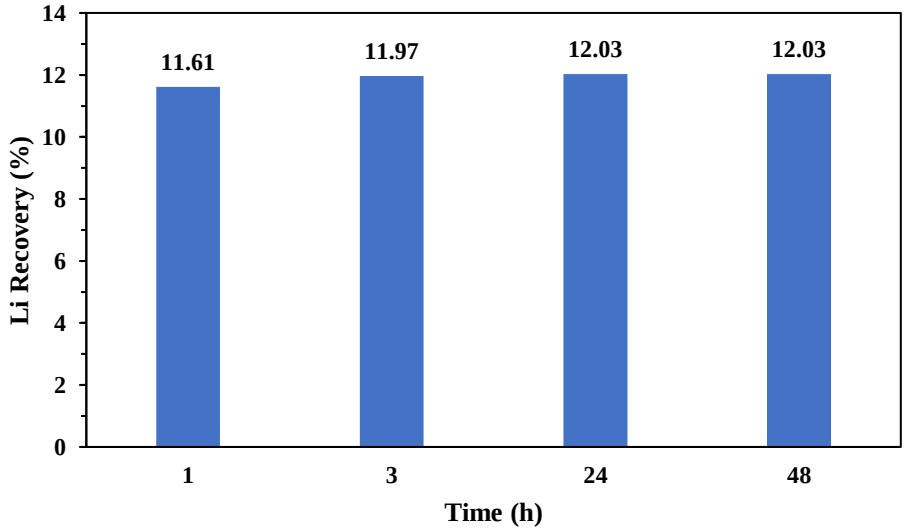


Figure S6. Neutral leaching results after thermal treatment of cathode powder in the presence of eggshell at 130 °C up to 48 hours (cathode/eggshell weight ratio of 2). Leaching conditions: room temperature, solid/liquid ratio of 2%, 1 hour of dissolution time.

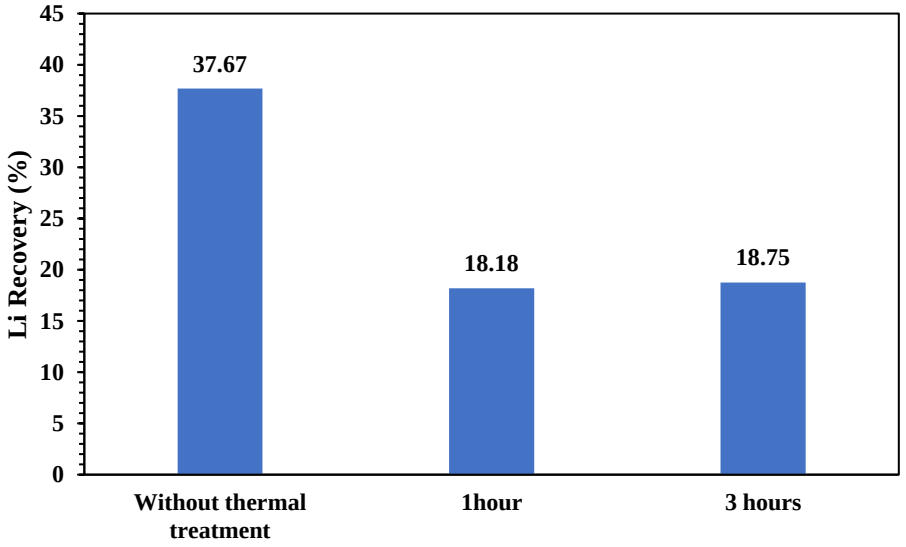


Figure S7. Neutral leaching results versus time of thermal treatment at 100 °C, for the milled samples of cathode powder for 8 hours, in the presence of eggshell (cathode/eggshell weight ratio of 4). Leaching conditions: room temperature, solid/liquid ratio of 2%, 1 hour of dissolution time.

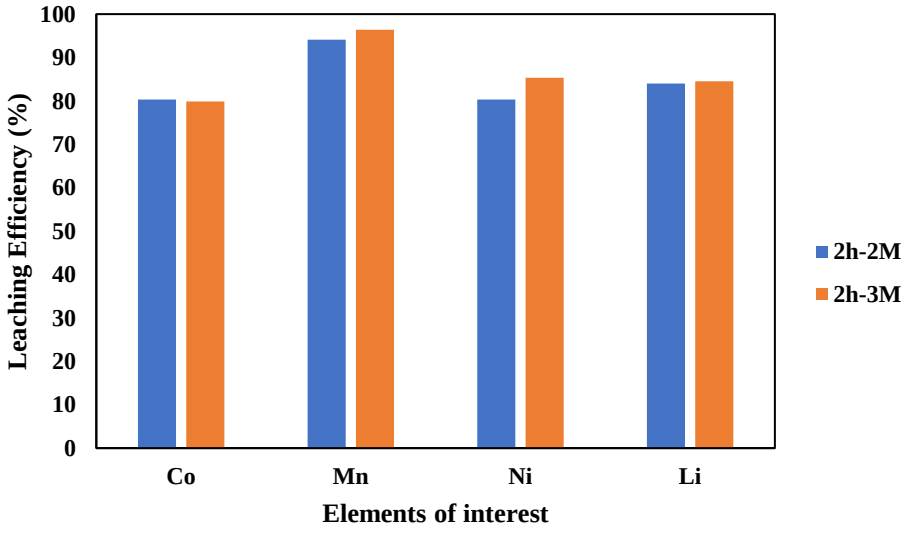


Figure S8. Effect of sulfuric acid concentration on the leaching efficiency of elements of interest at the highest hydrogen peroxide concentration (8%v). Leaching conditions: 8%V H_2O_2, 10% pulp density, and room temperature for 2 hours.

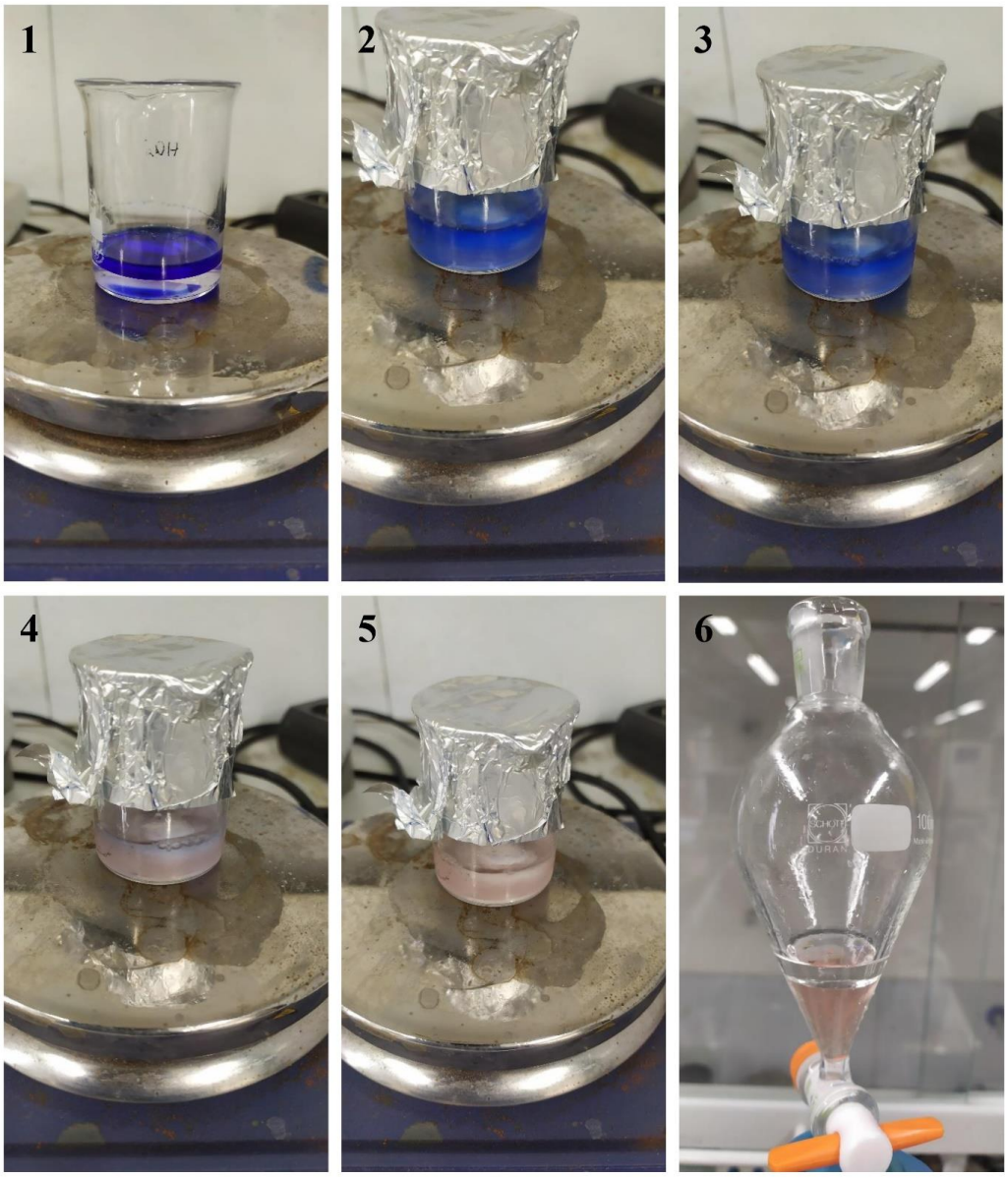


Figure S9. Color evolution of the organic and aqueous phases during the Co stripping stage.

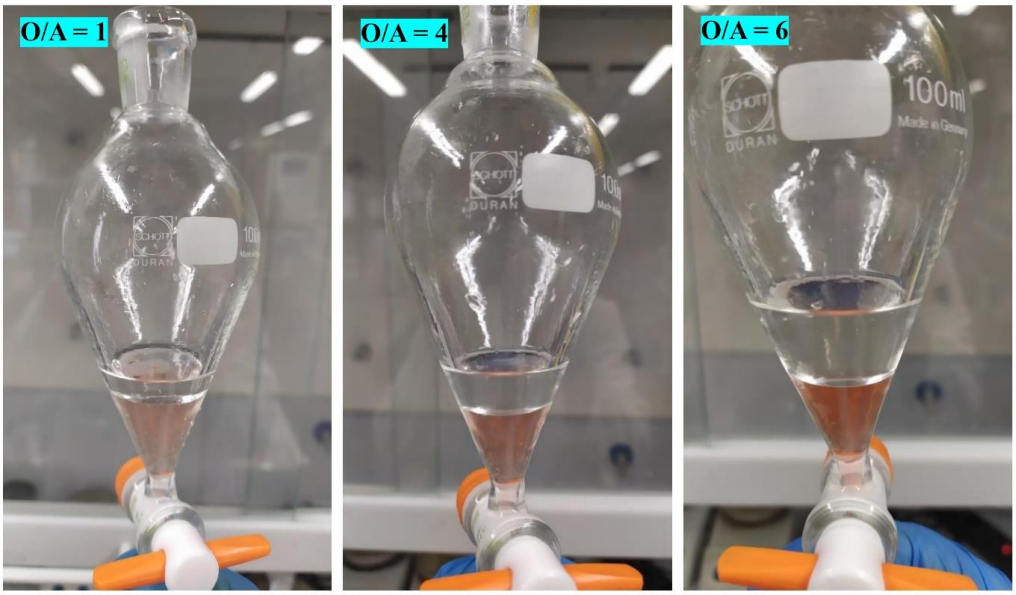


Figure S10. Phase separation after 5 minutes Co stripping with 0.5M sulfuric acid solution at various organic/aqueous phase ratios.

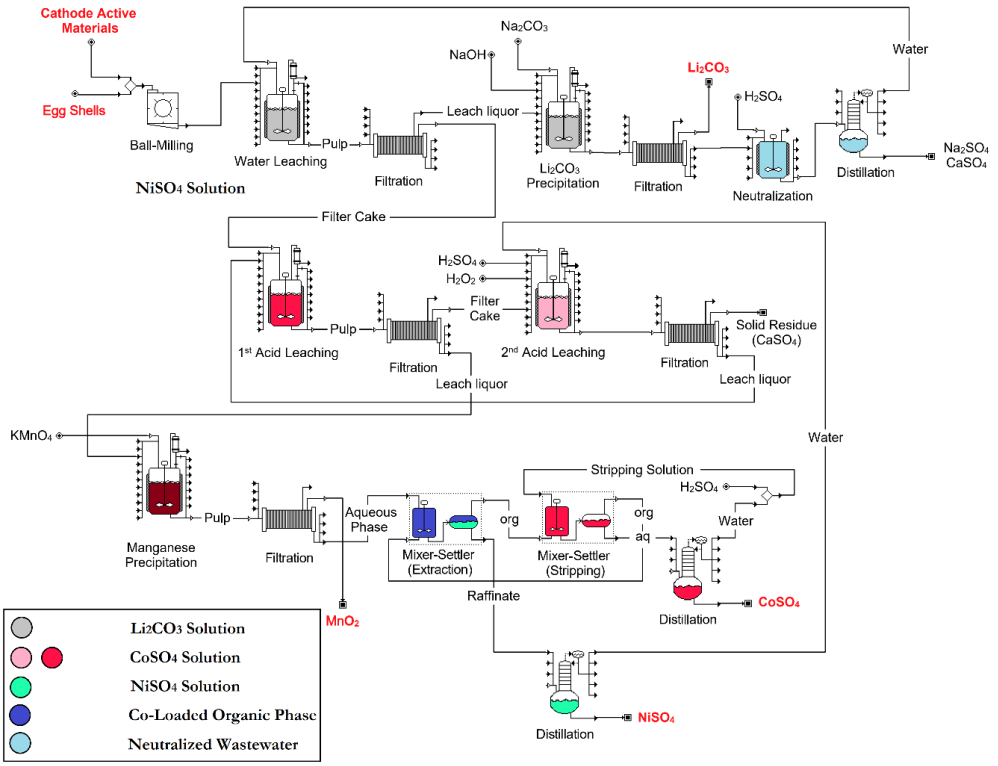


Figure S11. Process flow diagram for the potential scenario of selective hydrometallurgical recovery of lithium and the targeted elements from cathode-active materials of a mixture of spent Li-ion batteries.