Supporting Information

An Ultra-fast Reaction Process for Recycling Lithium Ion Batteries *via* Galvanic Cell Interaction

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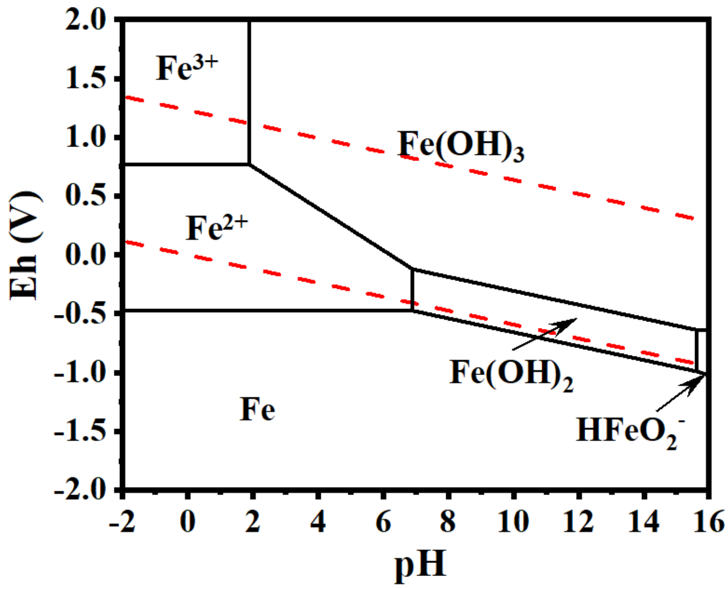


Figure S1. The Porbaix diagram of Fe-H_2O.

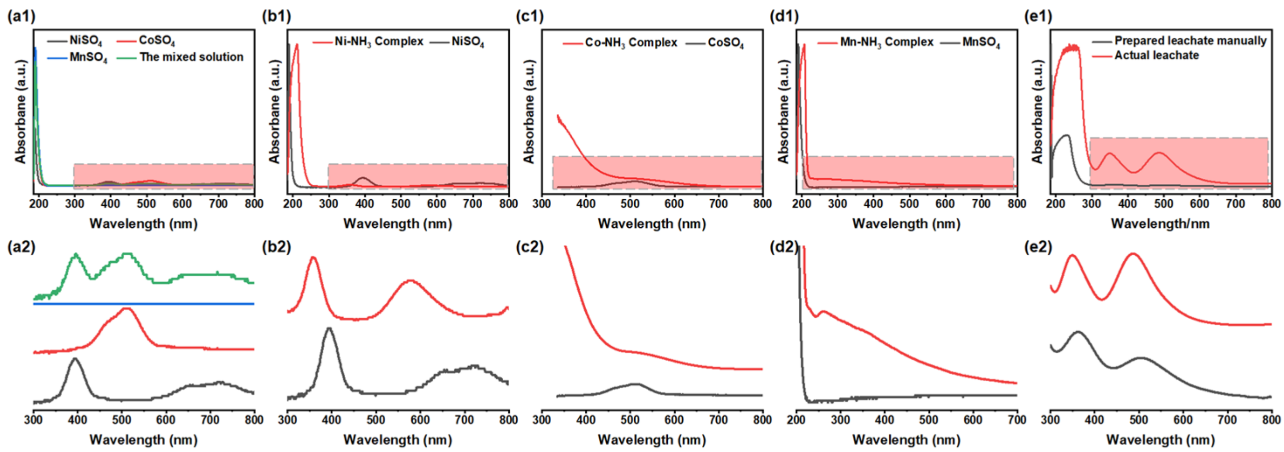


Figure S2. The UV-vis results of (a1-d1 and a2-d2) Ni^{2+}, Co^{2+}, Mn^{2+} in two medias of water and leaching reagent, (e1 and e2) the actual leachate and prepared leachate manually.

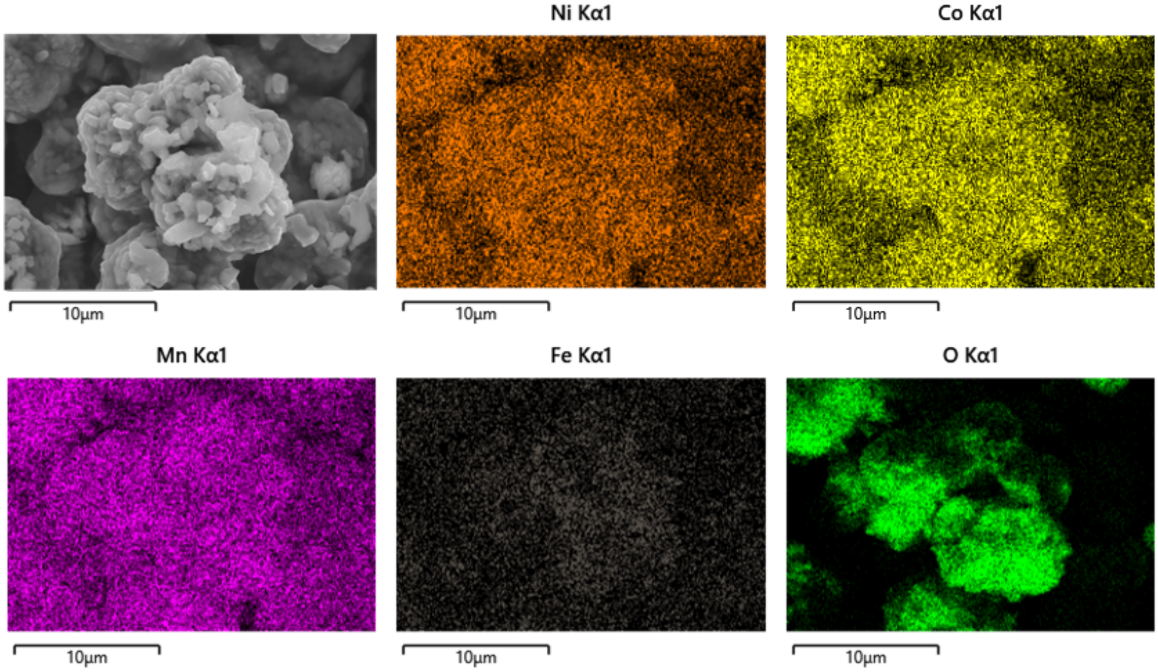


Figure S3. The EDS iamges for the NCM111 electrode after the external circuit detection test.

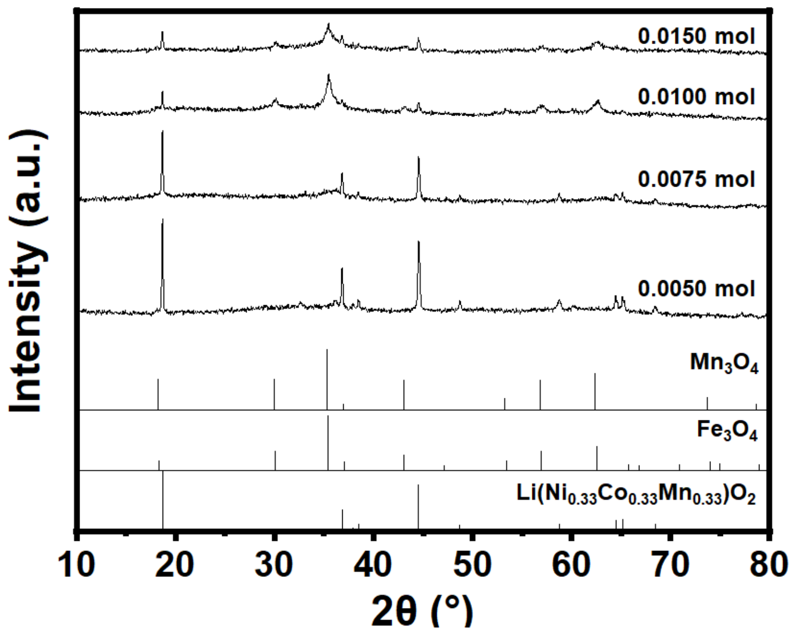


Figure S4. The XRD patterns of the leaching residues vary with a gradient of $FeSO_4$ usage.

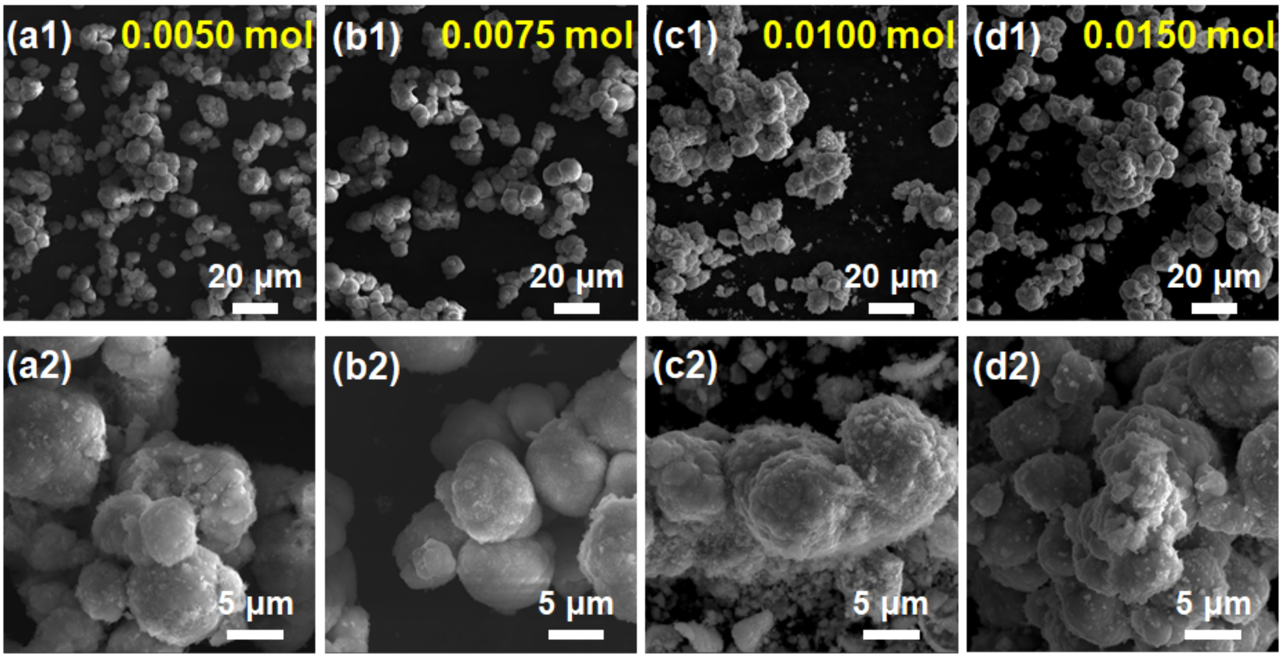


Figure S5. The SEM images of the leaching residues vary with a gradient of $FeSO_4$ usage.

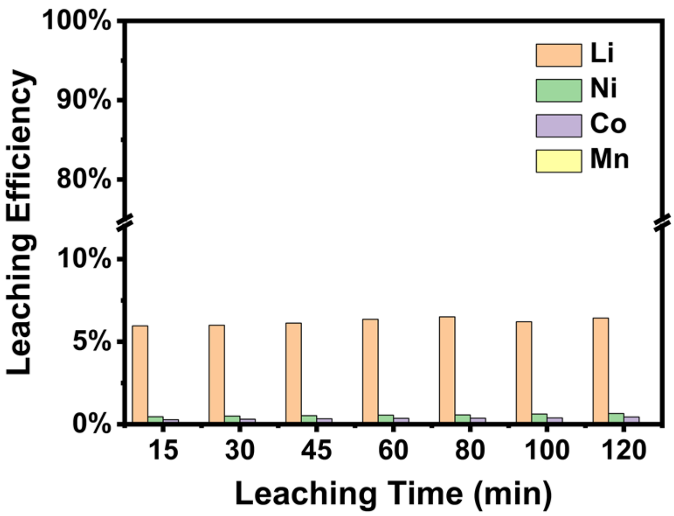


Figure S6. The leaching results without adding $FeSO_4$.

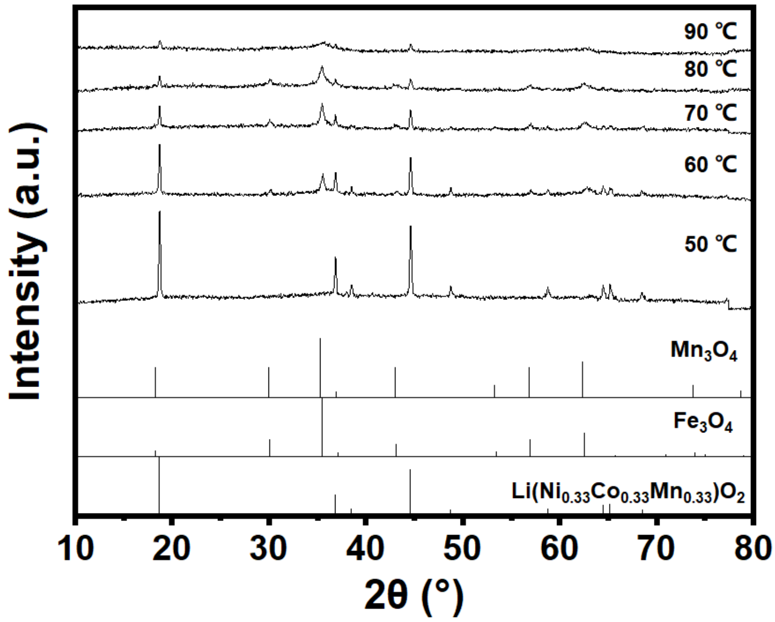


Figure S7. The XRD patterns of the leaching residues vary with a temperature gradient.

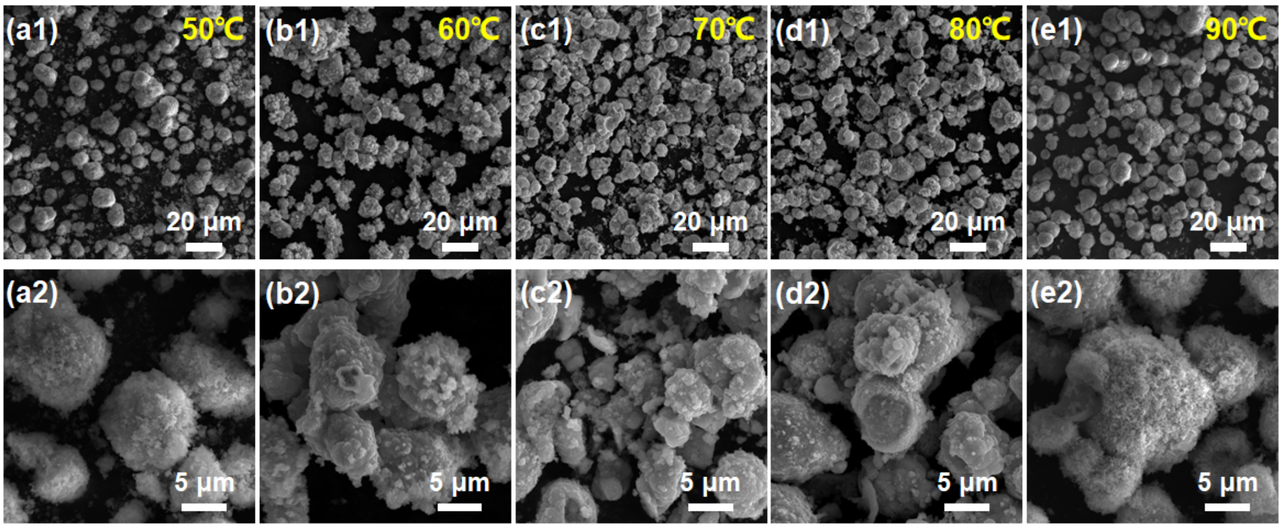
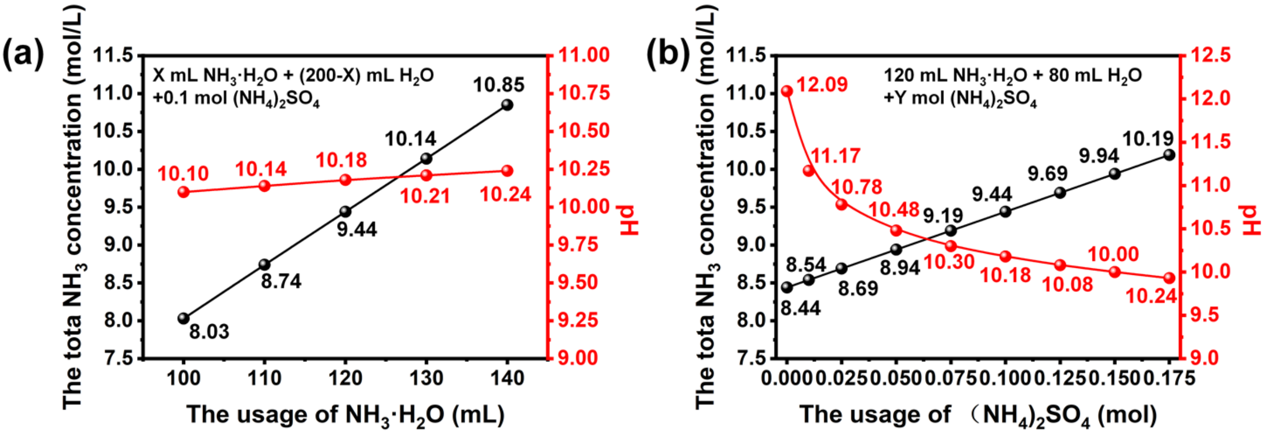


Figure S8. The SEM images of the leaching residues with a temperature gradient.



(c)

Agents	The usage amount				
$NH_3 \cdot H_2O$ (X mL)	30	45	60	80	120
H_2O (200-X mL)	170	155	140	120	80
$(NH_4)_2SO_4$ (Y mol)	0.025	0.038	0.05	0.067	0.1
Total NH_3 (mol/L)	2.36	3.54	4.72	6.29	9.44
pH	10.18	10.18	10.18	10.18	10.18

Figure S9. The changes of pH and total NH_3 concentration with elevated usage of (a) $NH_3 \cdot H_2O$ and (b) $(NH_4)_2SO_4$. (c) The changes as adjusting $NH_3 \cdot H_2O$ and $(NH_4)_2SO_4$ in euqal proportion.

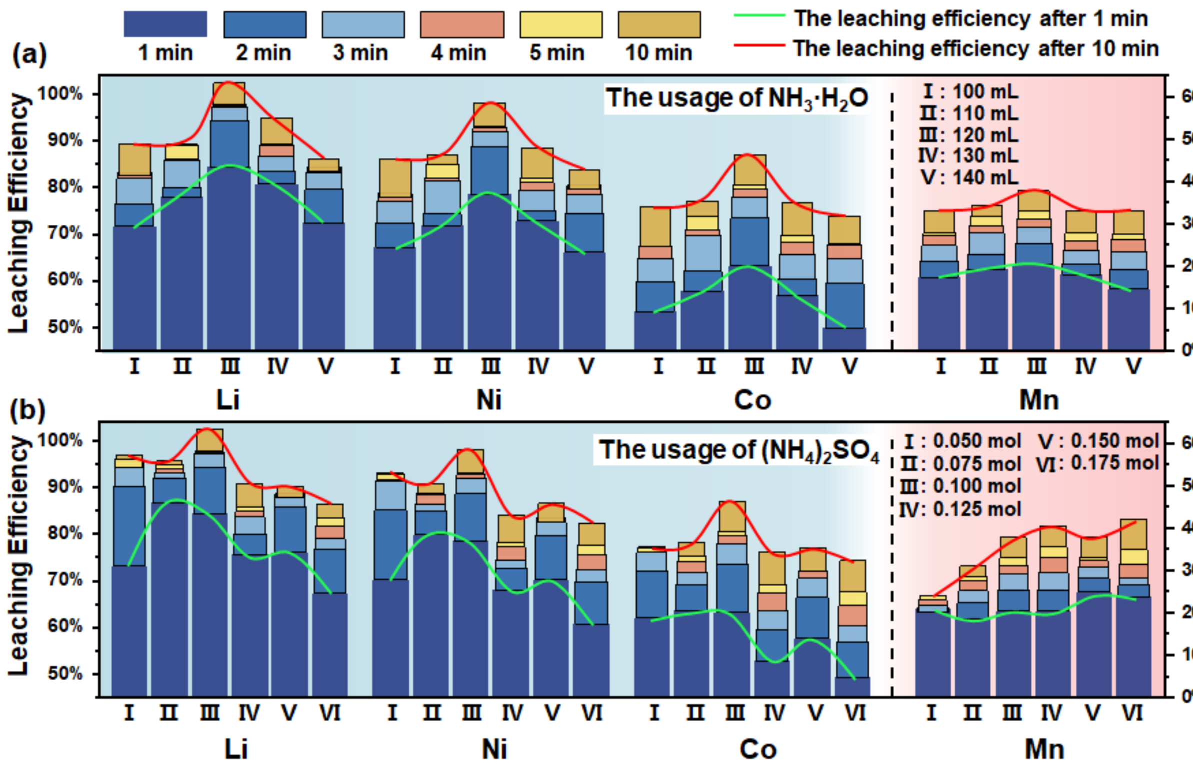


Figure S10. The leaching efficiency of valuable metals changes with the gradient of (a) $NH_3 \cdot H_2O$ usage, and (b) $(NH_4)_2SO_4$ usage.

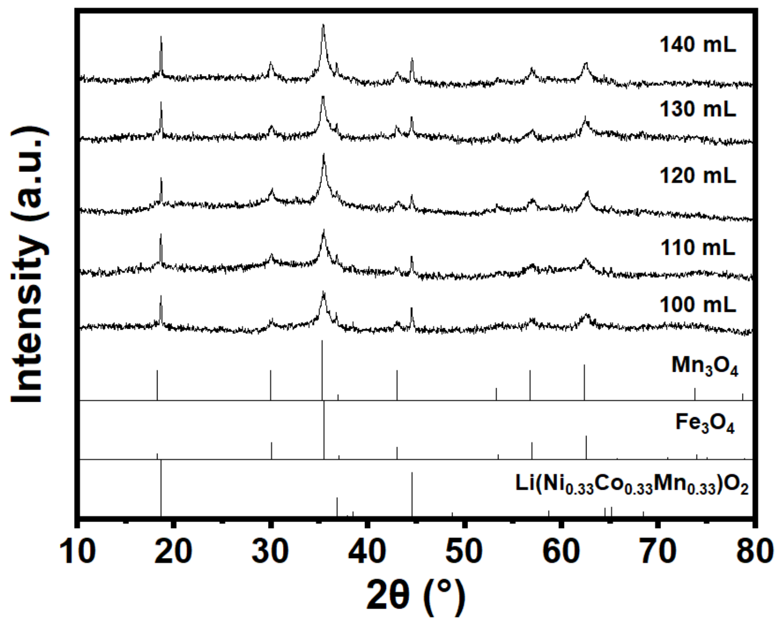


Figure S11. The XRD patterns of the leaching residues vary with a gradient of $NH_3 \cdot H_2O$ usage.

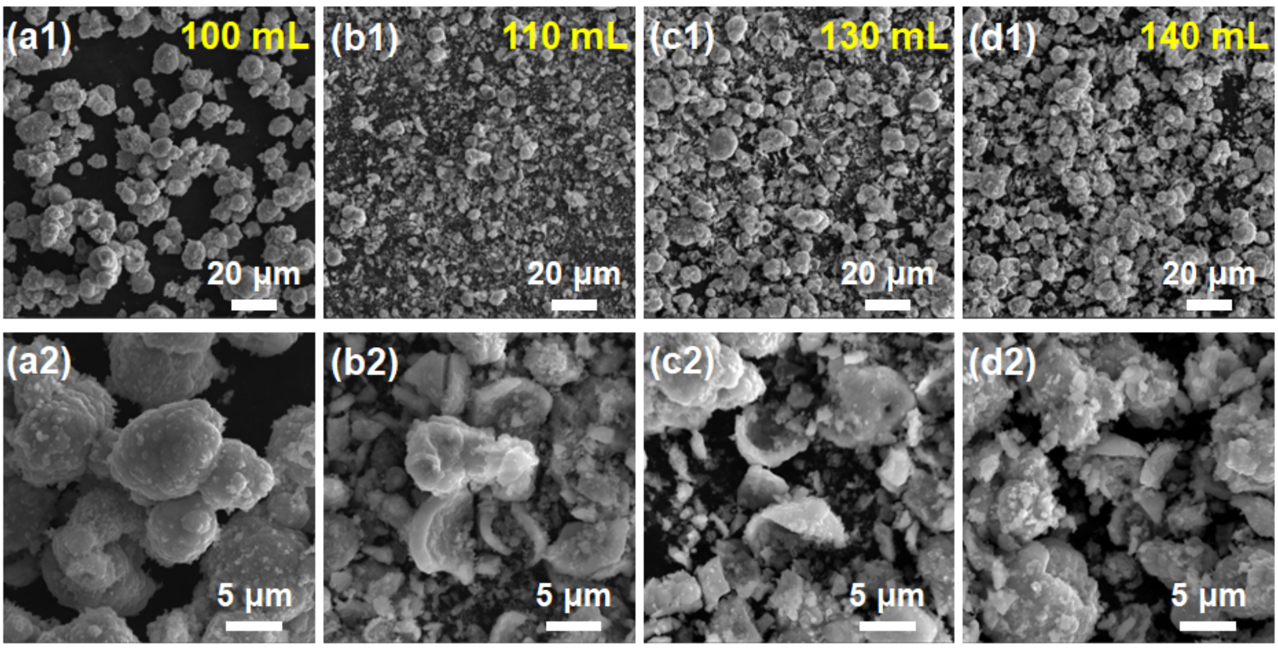


Figure S12. The SEM images of the leaching residues vary with a gradient of $NH_3 \cdot H_2O$ usage.

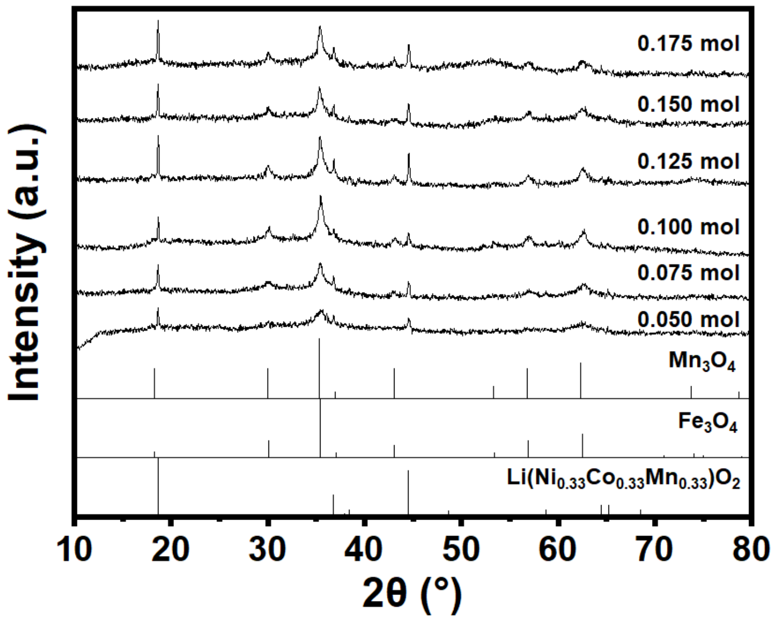


Figure S13. The XRD patterns of the leaching residues vary with a gradient of $(NH_4)_2SO_4$ usage.

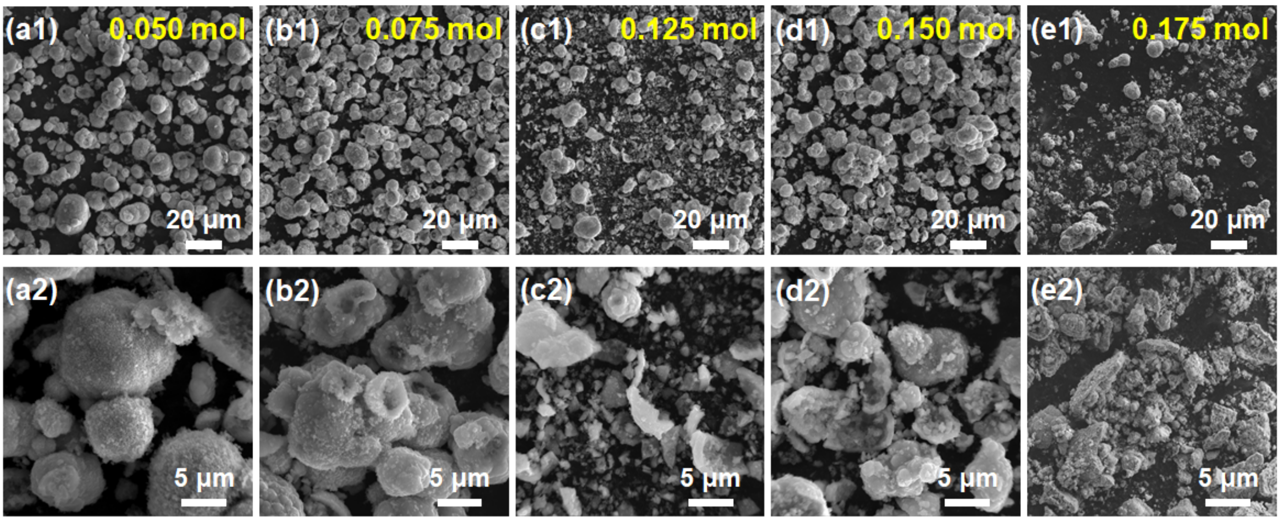


Figure S14. The SEM images of the leaching residues vary with a gradient of $(NH_4)_2SO_4$ usage.

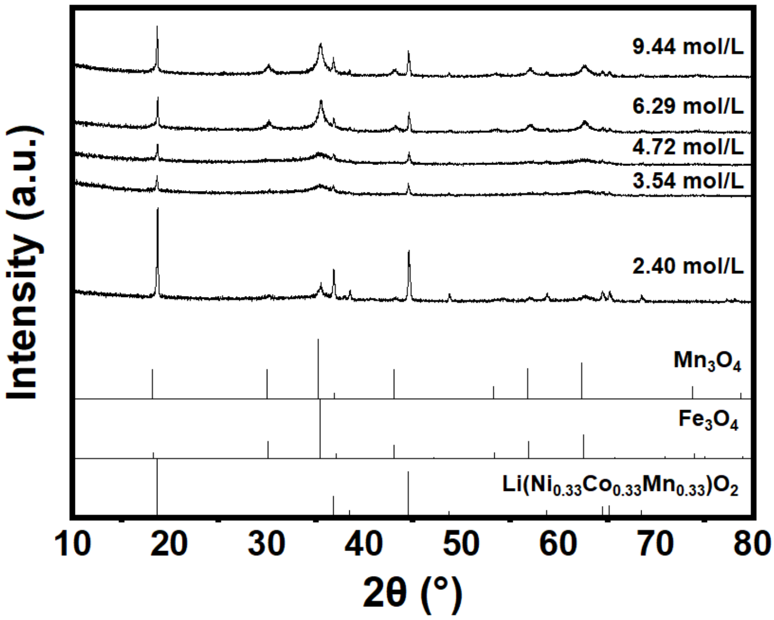


Figure S15. The XRD patterns of the leaching residues vary with a gradient in the concentration of NH_3.

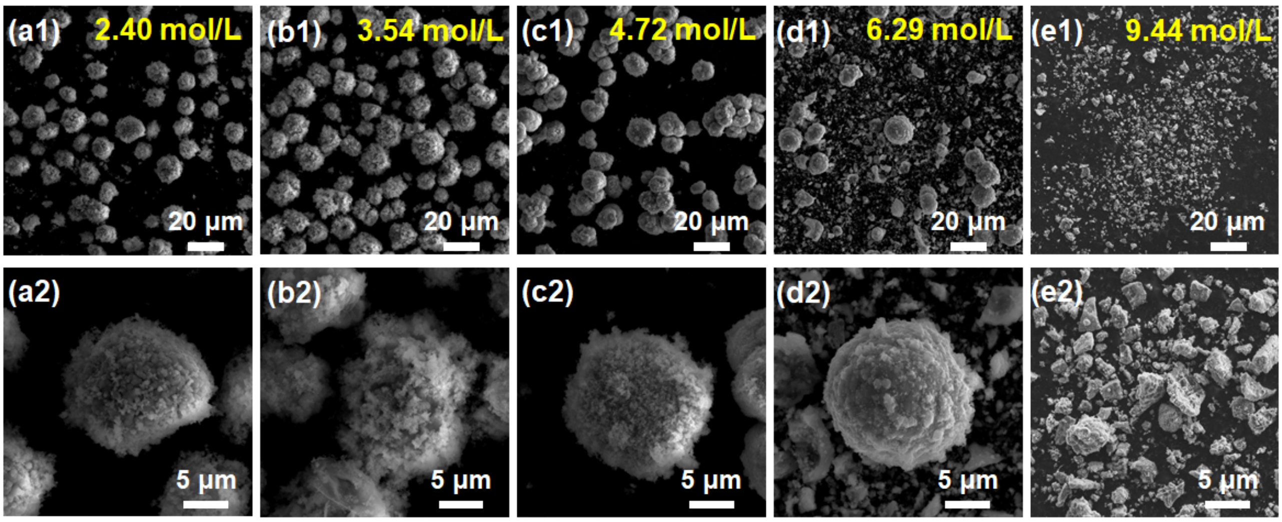


Figure S16. The SEM images of the leaching residues vary with a gradient in the concentration of NH_3.

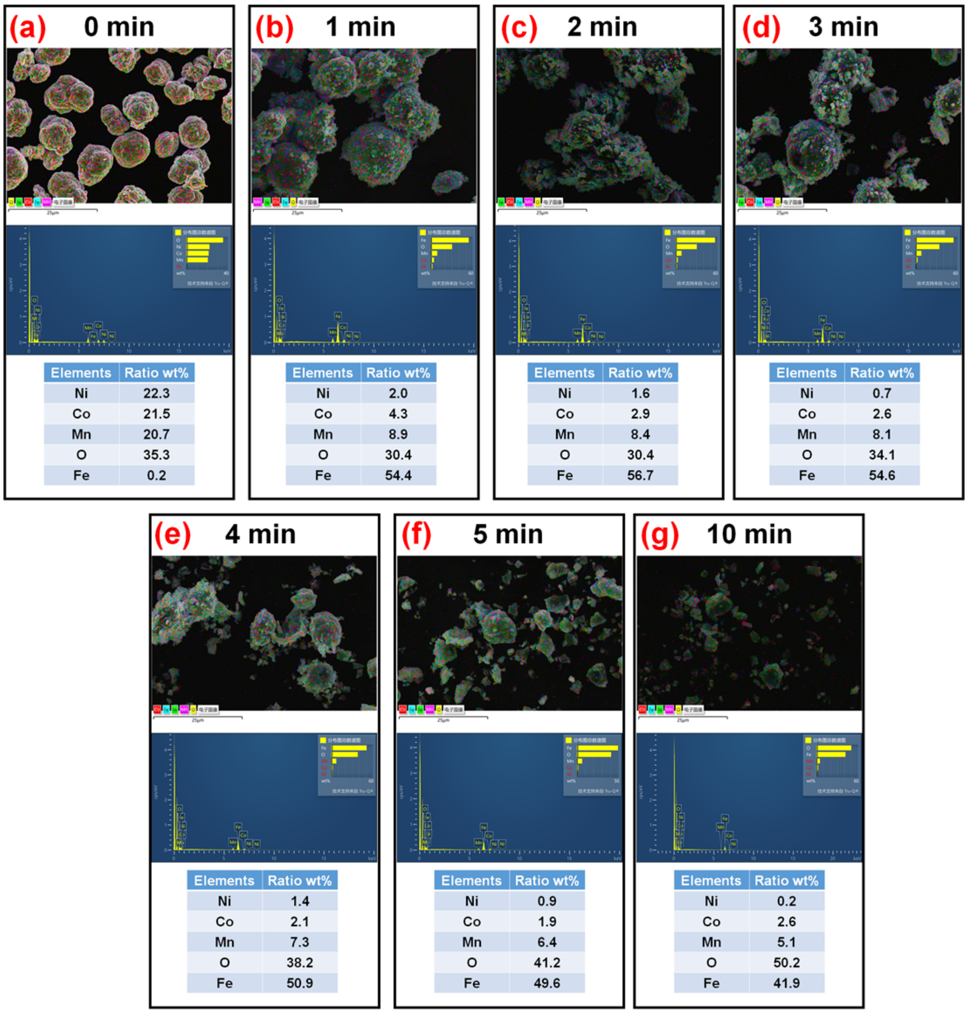


Figure S17. The EDS tests for the leaching residues at (a) 0 min, (b) 1 min, (c) 2 min, (d) 3 min, (e) 4 min, (f) 5 min, (g) 10 min.

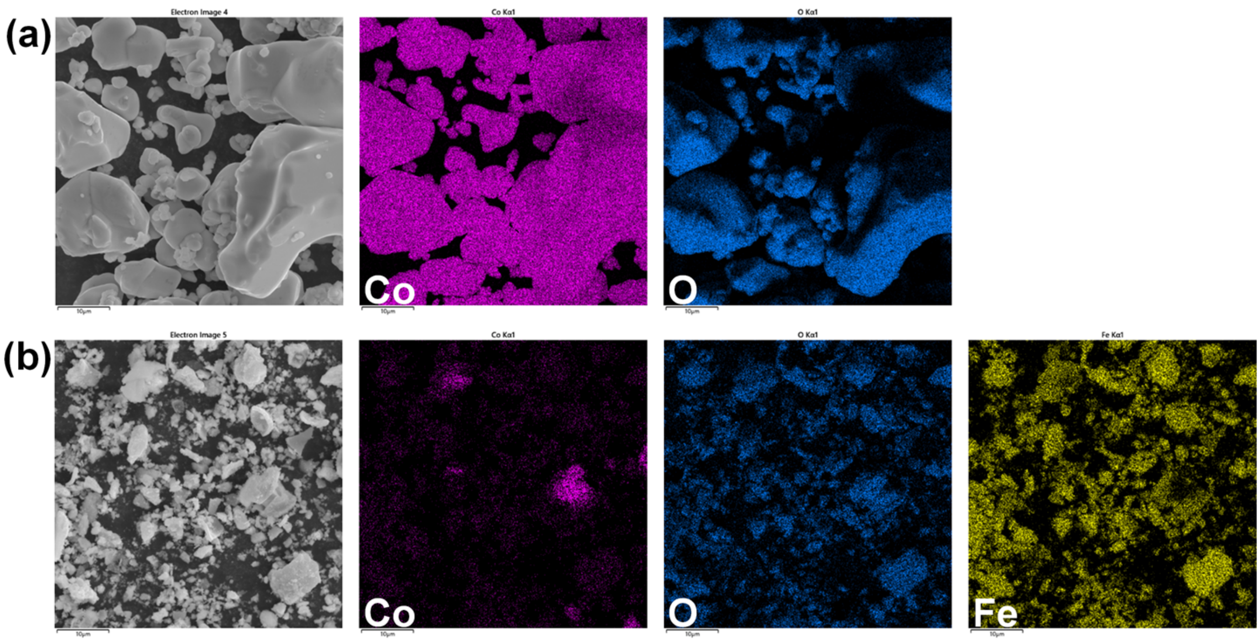


Figure S18. The EDS test of the LCO (a) before and (b) after leaching

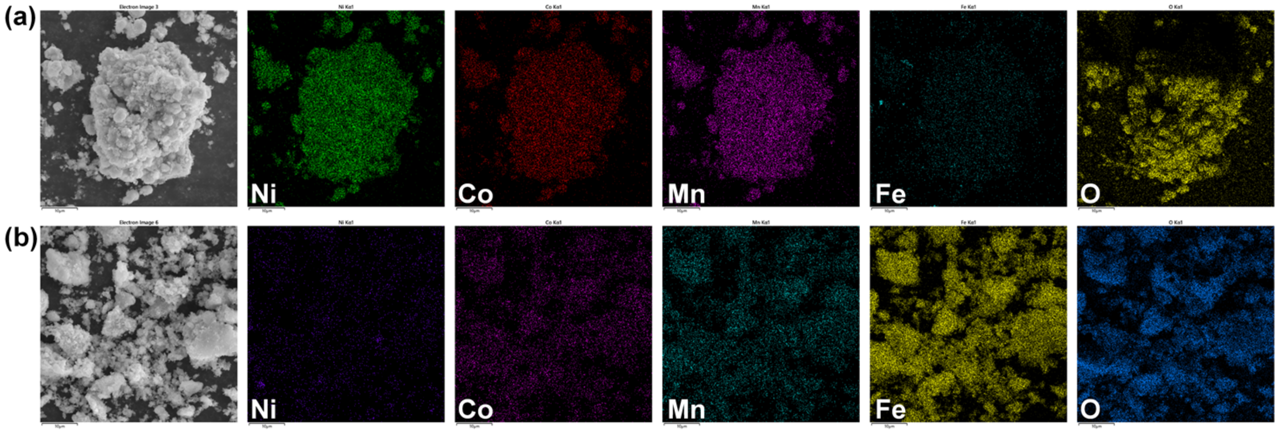


Figure S19. The EDS test of the NCM622 (a) before and (b) after leaching

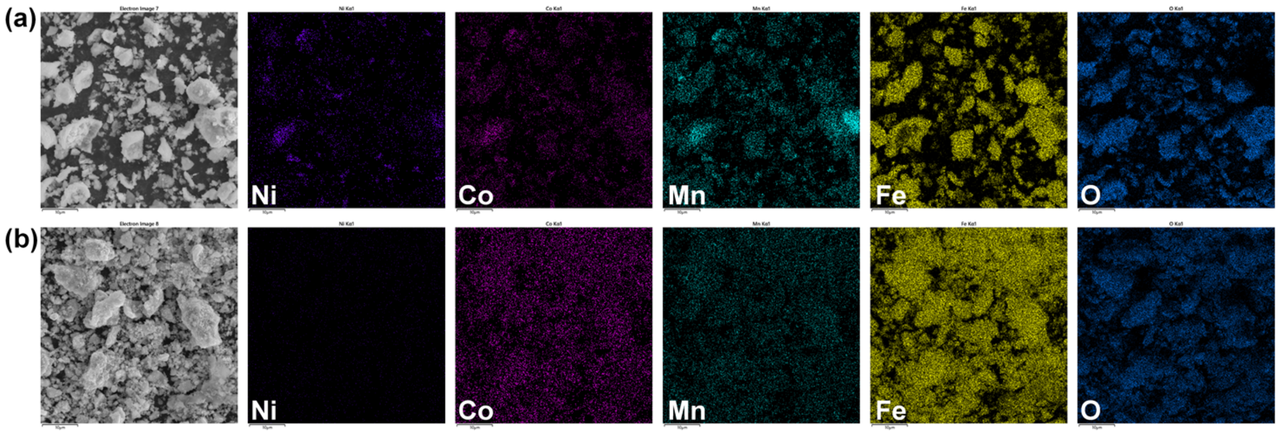


Figure S20. The EDS test of the two steps leaching: the (a) first and (b) second leaching residues

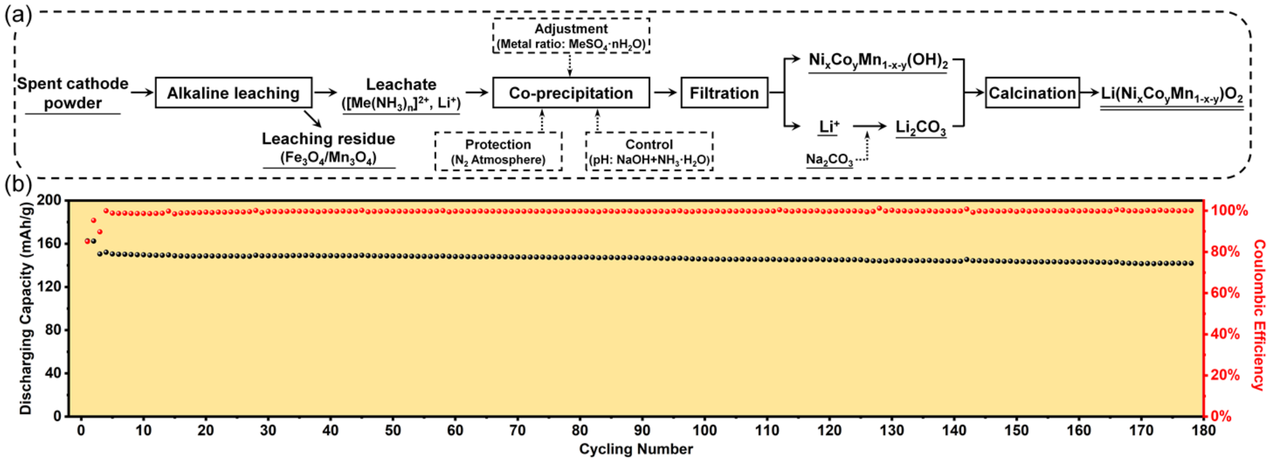


Figure S21. (a) The regeneration process. (b)The long-term cycling performance of regenerated NCM523 material

Table S1. The related information for Ni-H_2O Pourbaix diagram

Number	Chemical Equation	The calculated line when T = 298.15K
a	$2H^+ + 2e^- = H_2$	E = -0.059*pH
b	$O_2 + 4H^+ + 4e^- = 2H_2O$	E = 1.229 - 0.059*pH
1	$Ni^{3+} + e^- = Ni^{2+}$	E = 1.840
2	$Ni^{2+} + 2e^- = Ni$	E = -0.236
3	$Ni_2O_3 + 6H^+ = 2Ni^{3+} + 3H_2O$	pH = -0.646
4	$Ni(OH)_2 + 2H^+ = Ni^{2+} + 2H_2O$	pH = 6.357
5	$Ni_3O_4 + 2H_2O + 2H^+ + 2e^- = 3Ni(OH)_2$	E = 0.807 - 0.059*pH
6	$Ni_3O_4 + 8H^+ + 2e^- = 3Ni^{2+} + 4H_2O$	E = 1.935 - 0.237*pH
7	$Ni(OH)_2 + 2H^+ + 2e^- = Ni + 2H_2O$	E = 0.140 - 0.059*pH
8	$Ni_2O_3 + 6H^+ + 2e^- = 2Ni^{2+} + 3H_2O$	E = 1.725 - 0.178*pH
9	$3Ni_2O_3 + 2H^+ + 2e^- = 2Ni_3O_4 + H_2O$	E = 1.305 - 0.059*pH

Table S2. The related information for Co-H_2O Pourbaix diagram

Number	Chemical Equation	The calculated line when T = 298.15K
a	$2H^+ + 2e^- = H_2$	E = -0.059*pH
b	$O_2 + 4H^+ + 4e^- = 2H_2O$	E = 1.229 - 0.059*pH
1	$Co^{3+} + e^- = Co^{2+}$	E = 1.953
2	$Co^{2+} + 2e^- = Co$	E = -0.282
3	$Co_2O_3 + 6H^+ = 2Co^{3+} + 3H_2O$	pH = -3.348
4	$Co(OH)_2 + 2H^+ = Co2^+ + 2H_2O$	pH = 6.530
5	$Co_3O_4 + 2H_2O + 2H^+ + 2e^- = 3Co(OH)_2$	E = 0.485 - 0.059*pH
6	$Co_3O_4 + 8H^+ + 2e^- = 3Co^{2+} + 4H_2O$	E = 1.644 - 0.237*pH
7	$Co(OH)_2 + 2H^+ + 2e- = Co + 2H_2O$	E = 0.104 - 0.059*pH
8	$Co_2O_3 + 6H^+ + 2e^- = 2Co^{2+} + 3H_2O$	E = 1.358 - 0.178*pH
9	$3Co_2O_3 + 2H^+ + 2e^- = 2Co_3O_4 + H_2O$	E = 0.786 - 0.059*pH

Table S3. The related information for Mn-H_2O Pourbaix diagram

Number	Chemical Equation	The calculated line when T = 298.15K
a	$2H^+ + 2e^- = H_2$	E = -0.059*pH
b	$O_2 + 4H^+ + 4e^- = 2H_2O$	E = 1.229 - 0.059*pH
1	$Mn^{3+} + e^- = Mn^{2+}$	E = 1.542
2	$Mn^{2+} + 2e^- = Mn$	E = -1.182
3	$MnO_2 + 4H^+ + 2e^- = 2Mn^{2+} + 3H_2O$	E = 1.229 - 0.118*pH
4	$Mn(OH)_2 + 2H^+ = Mn^{2+} + 2H_2O$	pH = 7.282
5	$Mn_3O_4 + 2H_2O + 2H^+ + 2e^- = 3Mn(OH)_2$	E = 0.519- 0.059*pH
6	$Mn_3O_4 + 8H^+ + 2e^- = 3Mn^{2+} + 4H_2O$	E = 1.812 - 0.237*pH
7	$Mn(OH)_2 + 2H^+ + 2e^- = Mn + 2H_2O$	E = -0.751 - 0.059*pH
8	$2MnO_2 + 2H^+ + 2e^- = Mn_2O_3 + H_2O$	E = 0.974 - 0.059*pH
9	$Mn_2O_3 + 6H^+ + 2e^- = 2Mn^{2+} + 3H_2O$	E = 1.484 - 0.178*pH
10	$3Mn_2O_3 + 2H^+ + 2e^- = 2Mn_3O_4 + H_2O$	E = 0.829 - 0.059*pH
11	$MnO_4^- + e^- = MnO_4^{2-}$	E = 0.554
12	$MnO_4^- + 4H^+ + 3e^- = MnO_2 + 2H_2O$	E = 1.700 - 0.079*pH
13	$MnO_4^{2-} + 2e^- + 4H^+ = MnO_2 + 2H_2O$	E = 2.273 - 0.118*pH

Table S4. The related information for Fe-H_2O Pourbaix diagram

Number	Chemical Equation	The calculated line when T = 298.15K
a	$2H^+ + 2e^- = H_2$	E = -0.059*pH
b	$O_2 + 4H^+ + 4e^- = 2H_2O$	E = 1.229 - 0.059*pH
1	$Co^{3+} + e^- = Co^{2+}$	E = 1.953
2	$Co^{2+} + 2e^- = Co$	E = -0.282
3	$Co_2O_3 + 6H^+ = 2Co^{3+} + 3H_2O$	pH = -3.348
4	$Co(OH)_2 + 2H^+ = Co^{2+} + 2H_2O$	pH = 6.530
5	$Co_3O_4 + 2H_2O + 2H^+ + 2e^- = 3Co(OH)_2$	E = 0.485 - 0.059*pH
6	$Co_3O_4 + 8H^+ + 2e^- = 3Co^{2+} + 4H_2O$	E = 1.644 - 0.237*pH
7	$Co(OH)_2 + 2H^+ + 2e^- = Co + 2H_2O$	E = 0.104 - 0.059*pH
8	$Co_2O_3 + 6H^+ + 2e^- = 2Co^{2+} + 3H_2O$	E = 1.358 - 0.178*pH
9	$3Co_2O_3 + 2H^+ + 2e^- = 2Co_3O_4 + H_2O$	E = 0.786 - 0.059*pH

Table S5. The leaching efficiency of five independent experiment

Element	Time/min	Leaching efficiency					Average	SD
Li	1	0.801	0.84304	0.89804	0.77954	0.75744	0.81581	0.05578
	2	0.85976	0.94167	0.95961	0.84909	0.83756	0.88954	0.05668
	3	0.93056	0.97233	0.9572	0.89394	0.90057	0.93092	0.03426
	4	0.89009	0.97323	0.95906	0.94032	0.93412	0.93936	0.03159
	5	0.96797	0.97804	0.971	0.96319	0.96214	0.96847	0.00645
	10	0.97359	1.02312	0.9951	0.99958	1.00822	0.99992	0.01819
Ni	1	0.73245	0.78575	0.81633	0.68059	0.6533	0.73368	0.06860
	2	0.79886	0.88537	0.84698	0.74807	0.73169	0.80219	0.06488
	3	0.87943	0.9191	0.87063	0.79156	0.79213	0.85057	0.05663
	4	0.84483	0.92625	0.85717	0.8404	0.82571	0.85887	0.03931
	5	0.92792	0.92919	0.88466	0.86443	0.86185	0.89361	0.03310
	10	0.94858	0.98121	0.90292	0.90988	0.9068	0.92988	0.03407
Co	1	0.56258	0.63224	0.65772	0.55206	0.50508	0.58194	0.06214
	2	0.64869	0.73422	0.6957	0.62715	0.58869	0.65889	0.05716
	3	0.74073	0.77922	0.73037	0.67874	0.66041	0.71789	0.04816
	4	0.72521	0.79604	0.72648	0.73242	0.70154	0.73634	0.03540
	5	0.80229	0.80633	0.75881	0.76033	0.74305	0.77416	0.02838
	10	0.83893	0.87046	0.79254	0.81937	0.80303	0.82487	0.03094
Mn	1	0.18081	0.20421	0.22817	0.22234	0.18993	0.20509	0.02031
	2	0.2501	0.25336	0.25247	0.28675	0.26182	0.2609	0.01511
	3	0.32075	0.2899	0.28386	0.34473	0.33325	0.31450	0.02669
	4	0.32991	0.31156	0.29437	0.39923	0.37753	0.34252	0.04438
	5	0.37395	0.32914	0.3151	0.4266	0.41807	0.37257	0.05045
	10	0.38024	0.37762	0.34605	0.49074	0.4812	0.41517	0.06610
Fe	1	0.07277	0.12247	0.11102	0.14909	0.17007	0.12508	0.03722
	2	0.01803	0.03946	0.03205	0.07709	0.08575	0.05048	0.02944
	3	8.5E-04	0.01665	0.01305	0.03871	0.03841	0.02154	0.01661
	4	6.3E-05	0.00433	0.00548	0.01379	0.01344	0.00742	0.00600
	5	8.2E-05	0.00269	0.00328	0.00501	0.00329	0.00287	0.00178
	10	0.00209	-0.0019	0.00318	0.00144	0.0014	0.00125	0.00188