Supporting Information

Intelligent Metal Recovery from Spent Li-ion Batteries: Machine Learning Breaks the Barriers of Traditional Optimizations

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The dataset was collected from the following references about roasting integrated with water leaching for metal recovery from spent LIBs:

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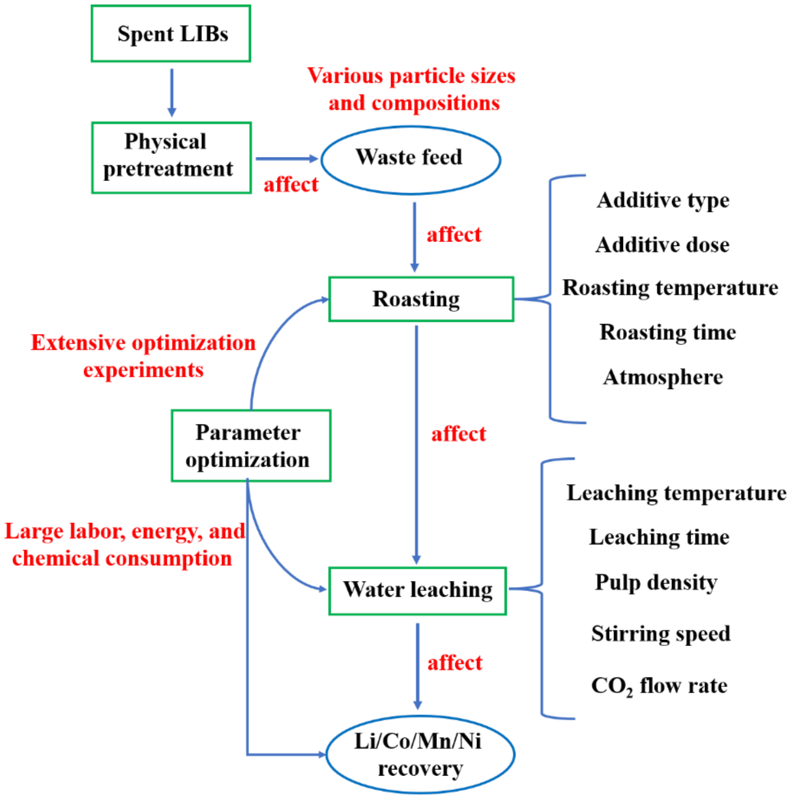

Fig. S1 The multiple influencing factors on the metal recovery from spent LIBs

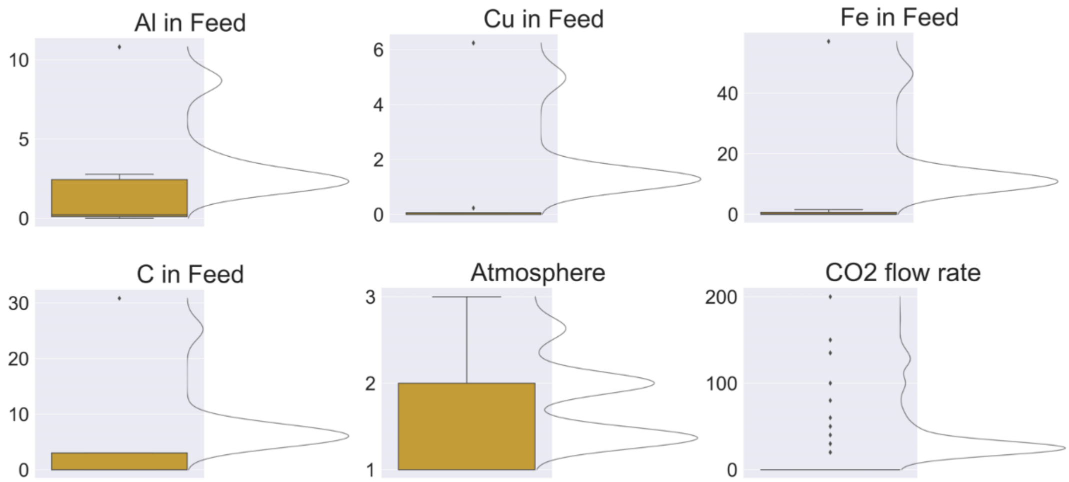

Figure S2 Box plots representing the descriptive statistics of the data for the other 6 features.

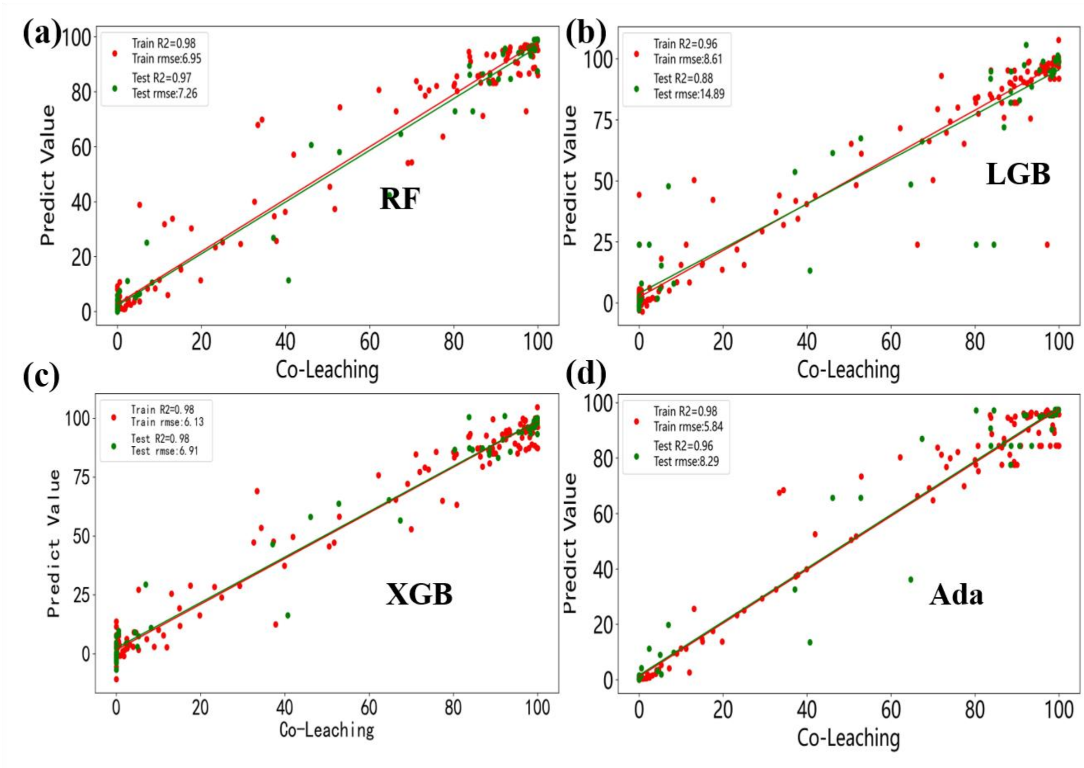

Figure S3 Scatter plot of predicted leaching efficiency of Co by four ML algorithms

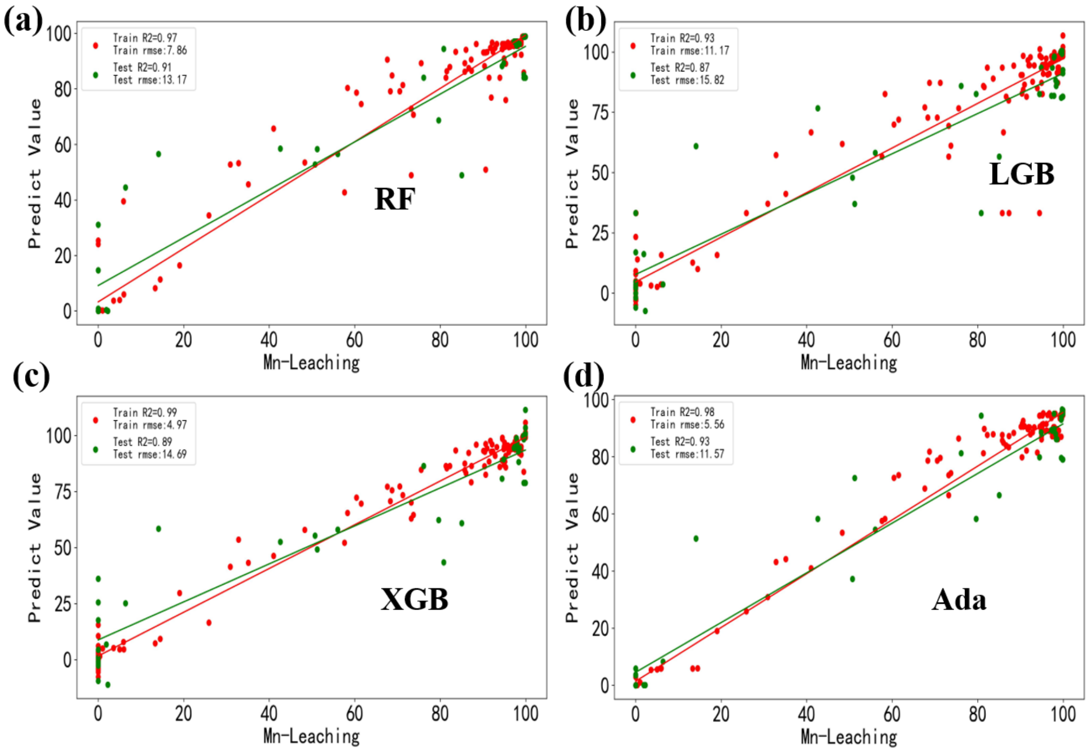

Figure S4 Scatter plot of predicted leaching efficiency of Mn by four ML algorithms

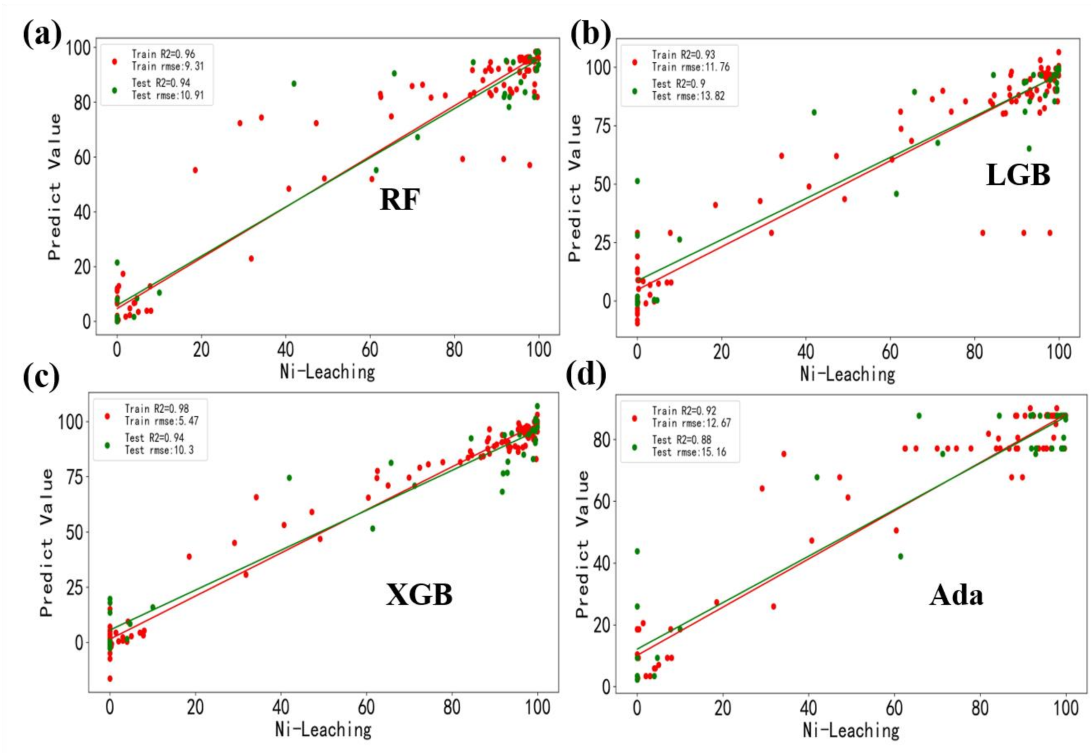

Figure S5 Scatter plot of predicted leaching efficiency of Ni by four ML algorithms

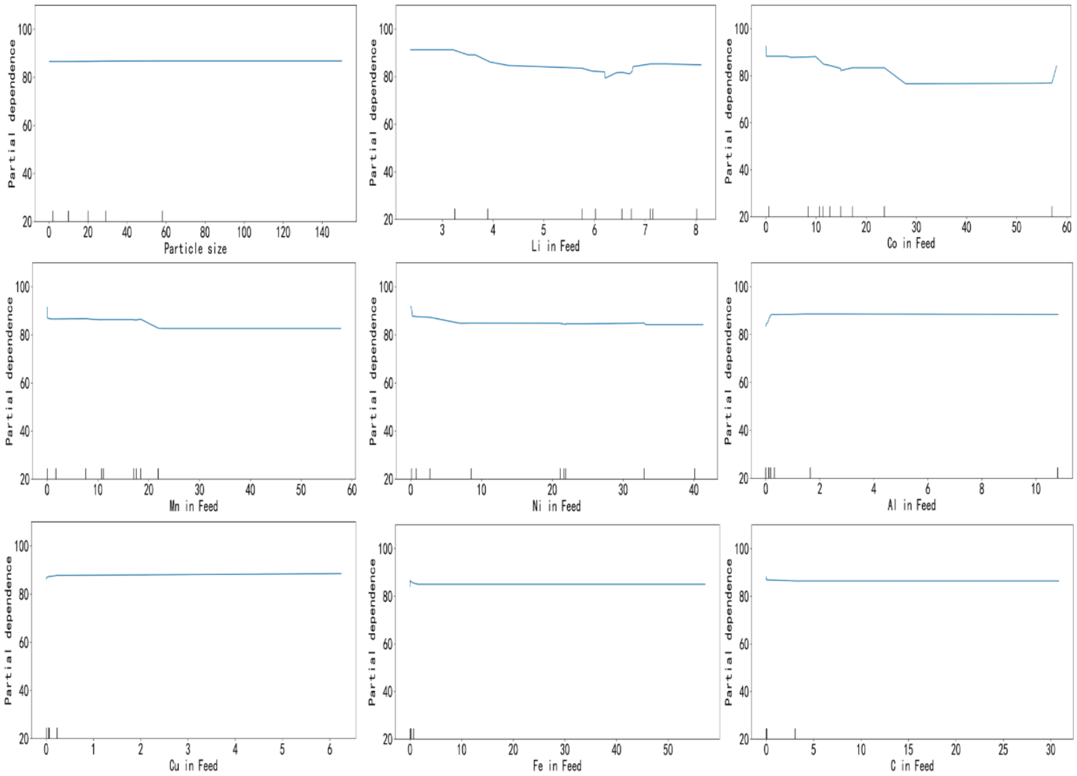

Figure S6 Partial dependence plots for waste property impacted Li leaching

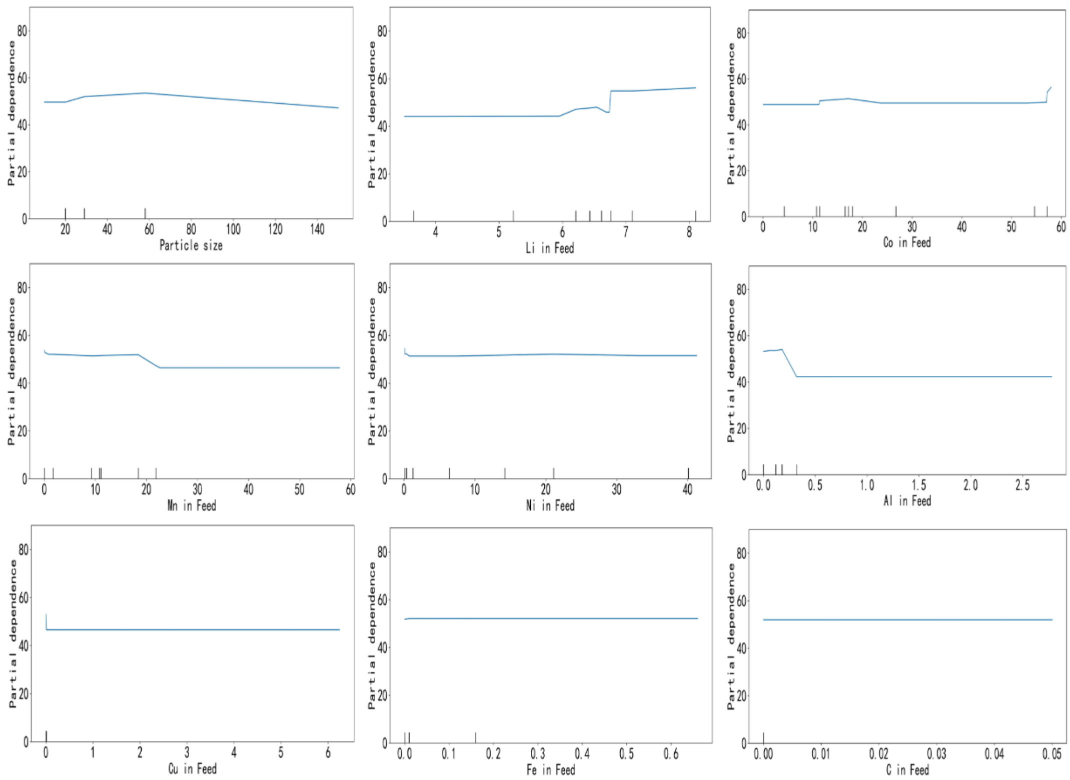

Figure S7 Partial dependence plots for waste property impacted Co leaching

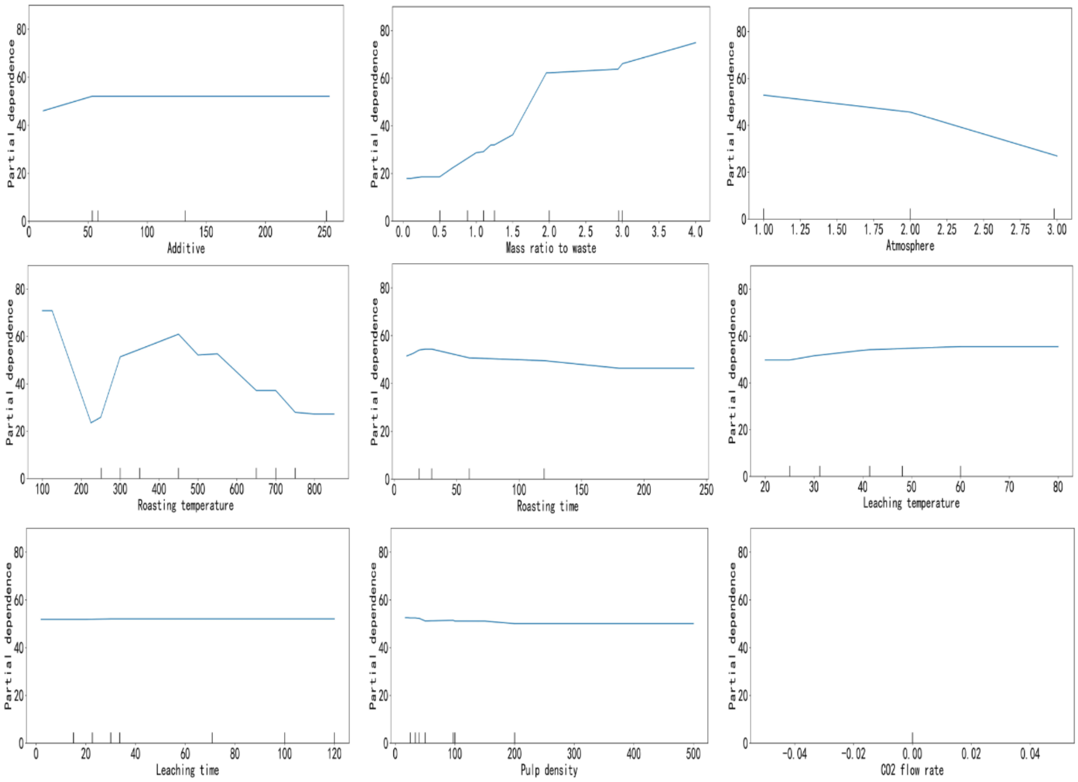

Figure S8 Partial dependence plots for roasting and water-leaching impacted Co leaching

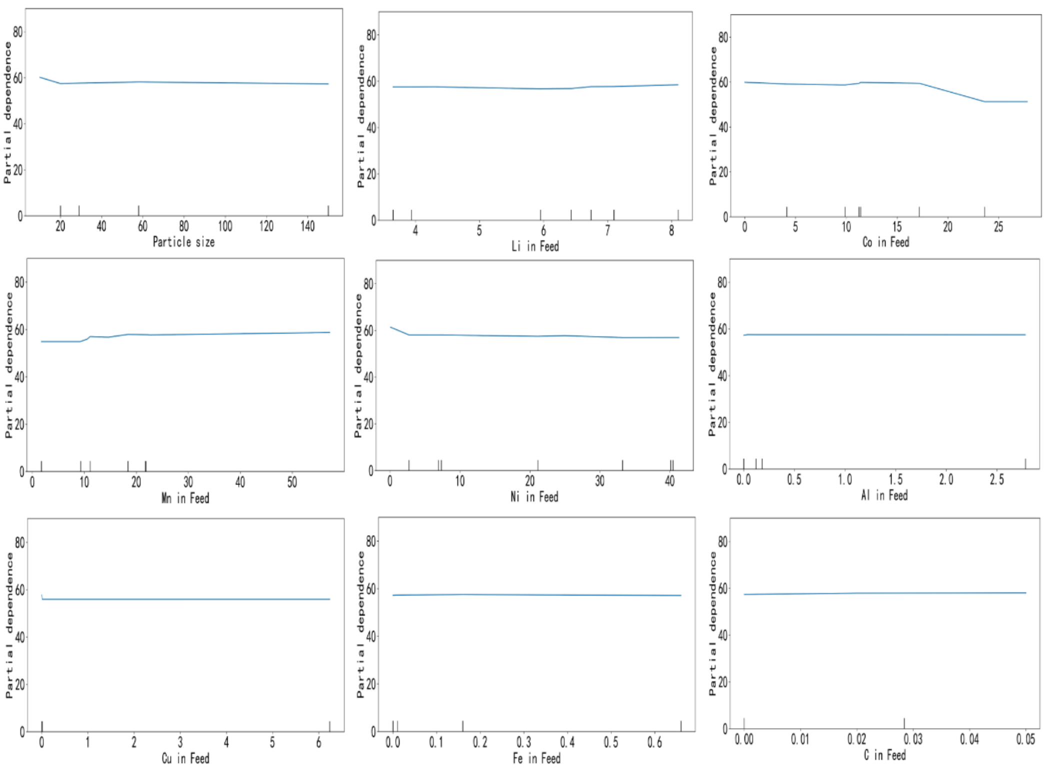

Figure S9 Partial dependence plots for waste property impacted Mn leaching

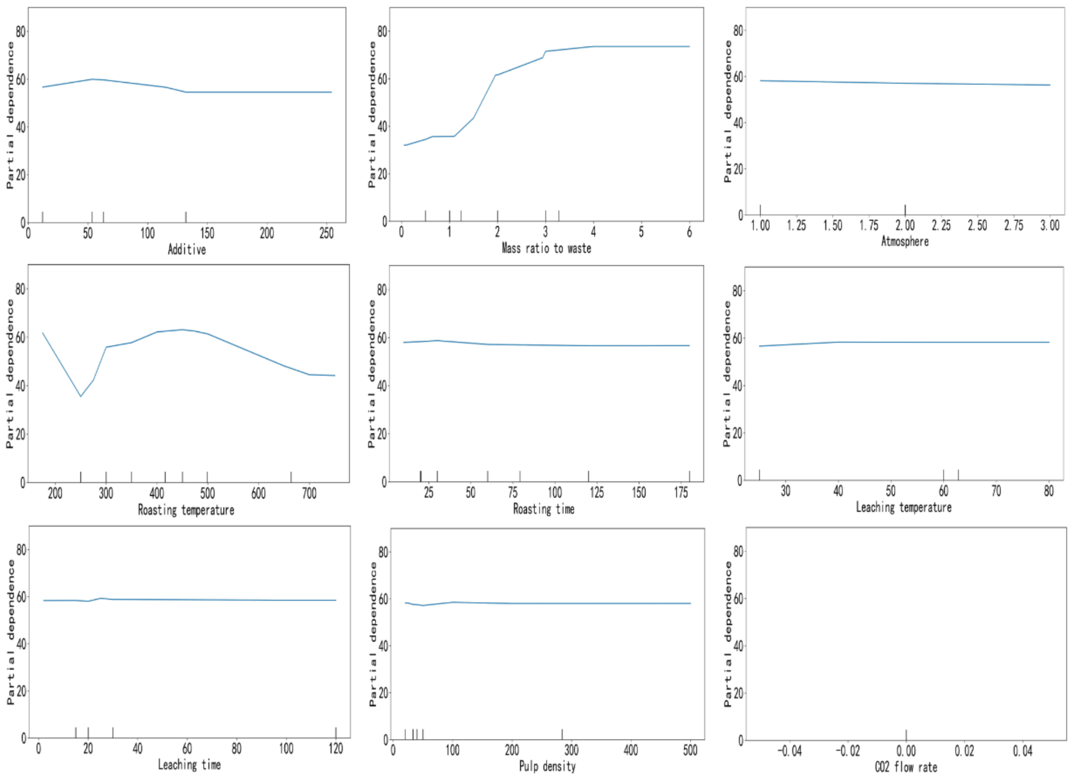

Figure S10 Partial dependence plots for roasting and water-leaching impacted Mn leaching

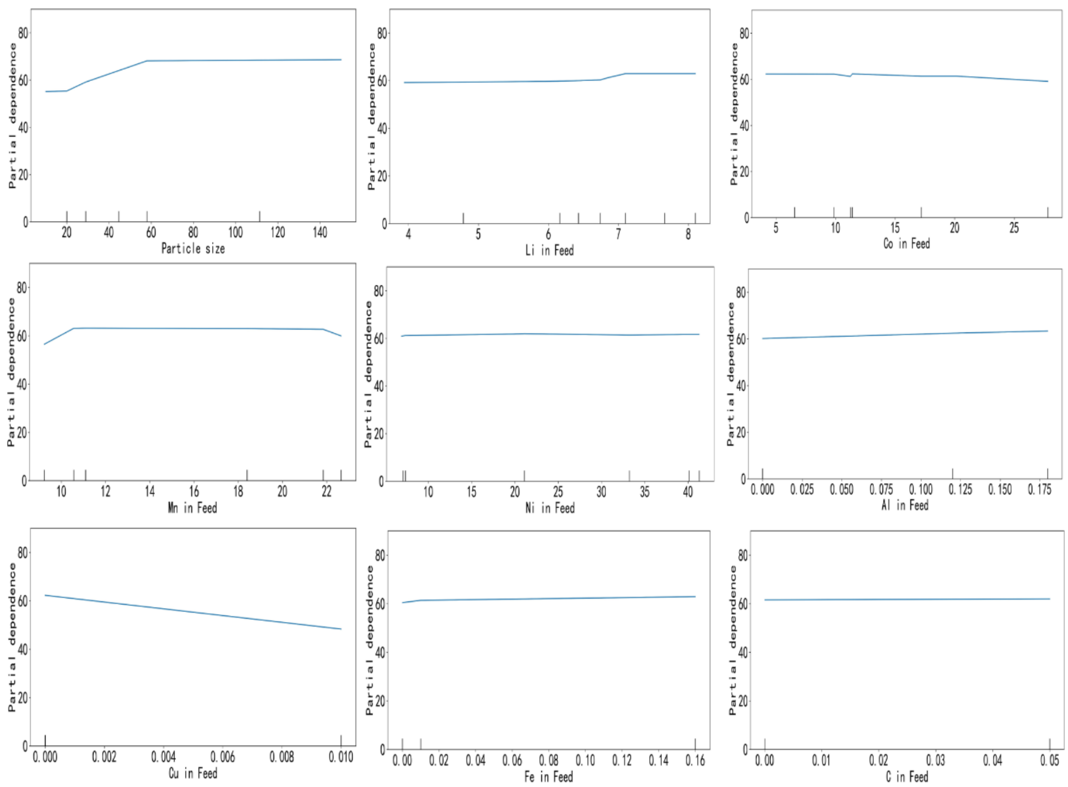

Figure S11 Partial dependence plots for waste property impacted Ni leaching

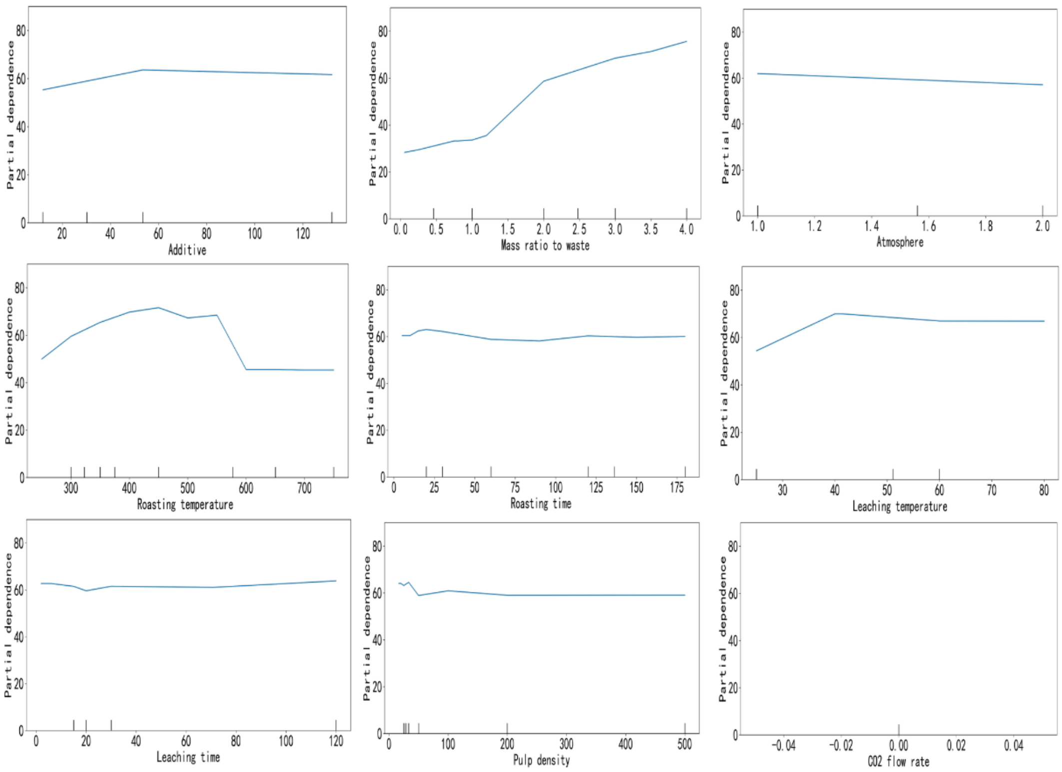

Figure S12 Partial dependence plots for roasting and water-leaching impacted Ni leaching

Table S1 The labeled values for the additives and roasting atmosphere used for machine learning

Additive type	Labeled values
NH_4Cl	53.5
NH_4HSO_4	115
$(NH_4)_2SO_4$	132
H_2SO_4	98
HNO_3	63
graphite	12
$Ca(OH)_2$	74
$K_2S_2O_7$	254
Zn	56
Lignite	1.1
Bean dregs	2
Atmosphere	
Air	1
Inert gas	2
Vacuum	3

Note: These labeled values do not impact the machine learning results but rather facilitate the identification of these input features.

Table S2 The compositions of waste feed

Cathode powder	Particle size	Composition (wt%)			
	(um)	Li	Co	Mn	Ni
1-$LiCoO_2$	~50	6.21	53.42	0.09	0.08
1-$LiMn_2O_4$	~50	3.96	0.08	48.7	0.06
1-$LiNi_xMn_yCo_zO_2$	~50	6.37	19.58	20.18	19.34
2-$LiCoO_2$	~50	6.41	52.35	-	-
	~100	6.36	51.83	-	-
2-$LiNi_xMn_yCo_zO_2$	~50	6.25	21.24	21.37	20.88
	~100	6.28	21.59	20.86	21.11
3-$LiCoO_2$	~50	6.73	57.15	-	-
	~100	6.69	56.97	-	-
3-$LiNi_xMn_yCo_zO_2$	~50	6.97	17.61	18.52	21.33
	~100	7.02	17.42	18.73	20.14

Note: the 1, 2 and 3 (such as 1- $LiCoO_2$, 2-$LiCoO_2$, 3-$LiCoO_2$ and 3-$LiNi_xMn_yCo_zO_2$)

repent different commercial LIBs from different companies.

Table S3 The experimental roasting and water-leaching parameters were set according to the GUI

Waste feed	Additive	Mass ratio of additive to waste	Roasting temperature (ºC)	Roasting time (min)	Atmosphere	Leaching temperature (ºC)	Leaching time (min)	Pulp density (g/L)	CO_2 flow rate (ml/min)
1-$LiCoO_2$(50 um)	graphite	0.25	700	180	vacuum	25	120	25	30
	NH_4Cl	4.0	350	30	air	25	30	150	0
1-$LiMn_2O_4$	graphite	0.25	700	30	vacuum	25	60	50	30
	NH_4Cl	3.6	350	30	air	25	30	150	0
1-$LiNi_xMn_yCoO_2$	graphite	0.25	750	180	vacuum	25	20	25	30
	NH_4Cl	3.8	350	25	air	25	120	160	0
2-$LiCoO_2$	graphite	0.28	700	150	vacuum	25	30	30	25.5
~100 um	graphite	0.28	700	150	vacuum	25	30	30	25.5
~100 um	NH_4Cl	4.0	300	20	air	25	30	170	0
2-$LiNi_xMn_yCo_zO_2$	graphite	0.5	680	100	vacuum	25	30	30	30
~100 um	NH_4Cl	3.8	375	30	air	25	30	200	0
3-$LiCoO_2$	graphite	0.25	700	100	vacuum	25	30	25	30
~100 um	NH_4Cl	4.0	320	20	air	30	20	250	0
3-$LiNi_xMn_yCo_zO_2$	graphite	0.5	680	100	vacuum	25	30	30	30
~100 um	NH_4Cl	3.5	300	30	air	30	30	250	0

Note: for the waste feed, the particle sizes are all ~50 um except the sample marked as (100) with ~100 um.

Table S4 The comparison with the traditional optimization and ML approach about the consumption of chemicals and electric energy and the production of waste water during

Optimizing parameters	Parameter values	NH_4Cl consumption (g)	Electric energy (kW•h)	Leaching waste water production (mL)
[a] NH_4Cl (g)	5	5	2	60
	10	10	2	60
	15	15	2	60
	20	20	2	60
	25	25	2	60
[b] Roasting temperature (°C)	300	20	2	60
	350	20	2	60
	400	20	2	60
	450	20	2	60
	500	20	2	60
[c] Roasting time (min)	10	20	0.7	60
	20	20	1.3	60
	30	20	2	60
	60	20	4	60
	90	20	6	60
Total	-	**275**	**34**	**900**
ML (only one-time experiment)	-	**20**	**1.3**	**52**

Note: we take the roasting of one type of spent $LiCoO_2$ (5 g) with NH_4Cl followed by water leaching as a typical example. The $LiCoO_2$ was chosen as the sample in **Table S2** of 2-$LiCoO_2$ with the particle size ~100 um. During the traditional optimization, we considered the NH_4Cl additive amount, roasting temperature, and roasting time as the required optimizing parameters, while the water leaching process was controlled as the leching temperature of 25 °C, pulp density of 150 g/L, and leaching time of 30 min. The power of the Muffle furnace used for the roasting process was 4 kW. The marked blue superscripts a, b, and c represent controlling other parameter values. **a**: roasting temperature of 400 °C, roasting time of 30 min; **b**: NH_4Cl additive amount of 20 g, roasting time of 30 min; **c**: roasting temperature of 300 °C, NH_4Cl additive amount of 20 g. After the roasting process under different conditions (as listed in **Table S3**), the mass of products (LiCl, $CoCl_2$, and residual $LiCoO_2$) was about 6~8.8 g. Then, the water-leaching process was conducted. Since the pulp density of 150 g/L, the added H_2O amount was 40~60 mL. Since most of the mass of products after roasting was distributed around 8.8 g, we chose the water-adding amount as 60 mL. Therefore, after each water leaching, the production amount of wastewater was about 60 mL. The GUI based on machine learning provided the parterres of NH_4Cl additive amount of 20 g, roasting temperature of 300 °C, roasting time of 20 min, leching temperature of 25 °C, pulp density of 170 g/L and leaching time of 30 min.