Supplementary Data

Using Sulfuric Acid–Phosphoric Acid Leaching to Remove Aluminum and Cathode Material Metal Residues from Spent Graphite

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**Table S1.** Removal (%) of studied elements ($n = 2$) shown in Figure 1a.

H$_3$PO$_4$ (mol/L)	Al (% ± %)	Co (% ± %)	Li (% ± %)	Mn (% ± %)	Ni (% ± %)
0	17.8 ± 1.1	98.9 ± 2.7	97.5 ± 0.6	98.3 ± 3.1	98.4 ± 2.9
0.1	30.2 ± 0.6	96.2 ± 0.8	92.9 ± 1.8	96.0 ± 1.0	96.0 ± 1.0
0.3	52.7 ± 2.4	95.6 ± 0.9	91.9 ± 0.1	95.5 ± 1.0	95.3 ± 0.8
0.5	56.8 ± 3.7	92.4 ± 0.3	91.1 ± 2.1	92.0 ± 0.3	92.3 ± 0.3
0.7	67.2 ± 0.6	93.4 ± 0.3	93.0 ± 2.6	92.9 ± 0.4	92.8 ± 0.6

Table S2. Removal (%) of studied elements ($n = 2$) shown in Figure 1b.

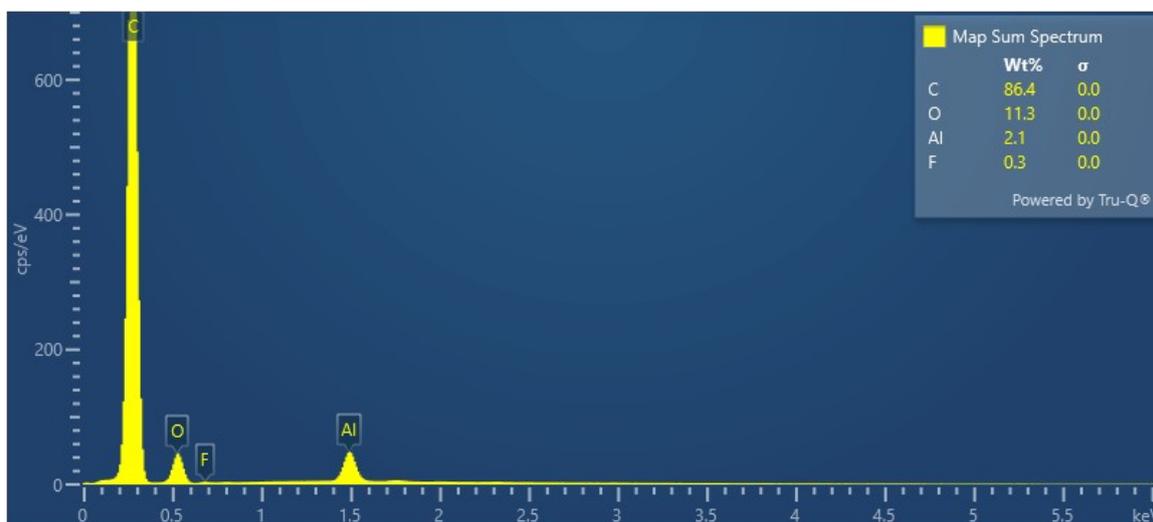| Time (min)	Al (% ± %)	Co (% ± %)	Li (% ± %)	Mn (% ± %)	Ni (% ± %)
240	56.8 ± 3.7	92.4 ± 0.3	91.1 ± 2.1	92.0 ± 0.3	92.3 ± 0.3
360	63.5 ± 2.5	97.3 ± 0.7	94.0 ± 2.2	97.2 ± 0.5	97.0 ± 1.0
480	80.5 ± 1.3	94.5 ± 0.8	94.3 ± 2.5	94.2 ± 0.0	93.9 ± 0.5
600	90.8 ± 0.6	99.2 ± 0.7	97.7 ± 0.7	99.5 ± 0.9	98.8 ± 0.7
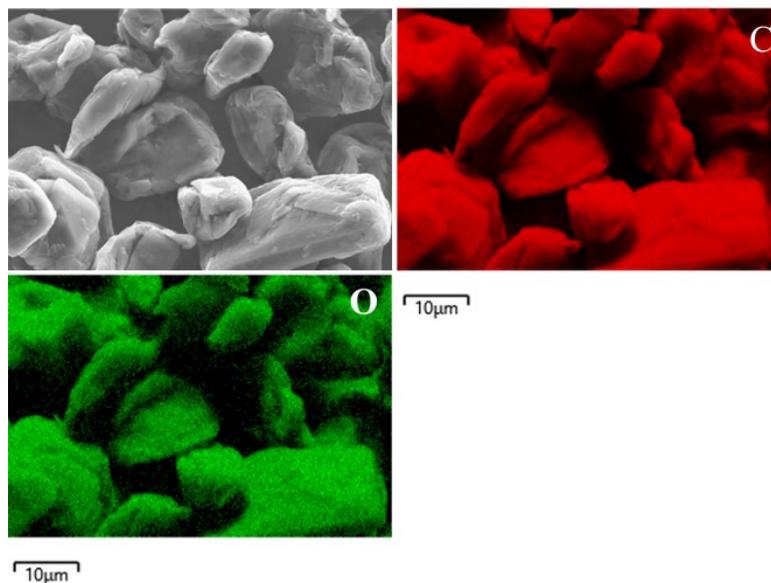
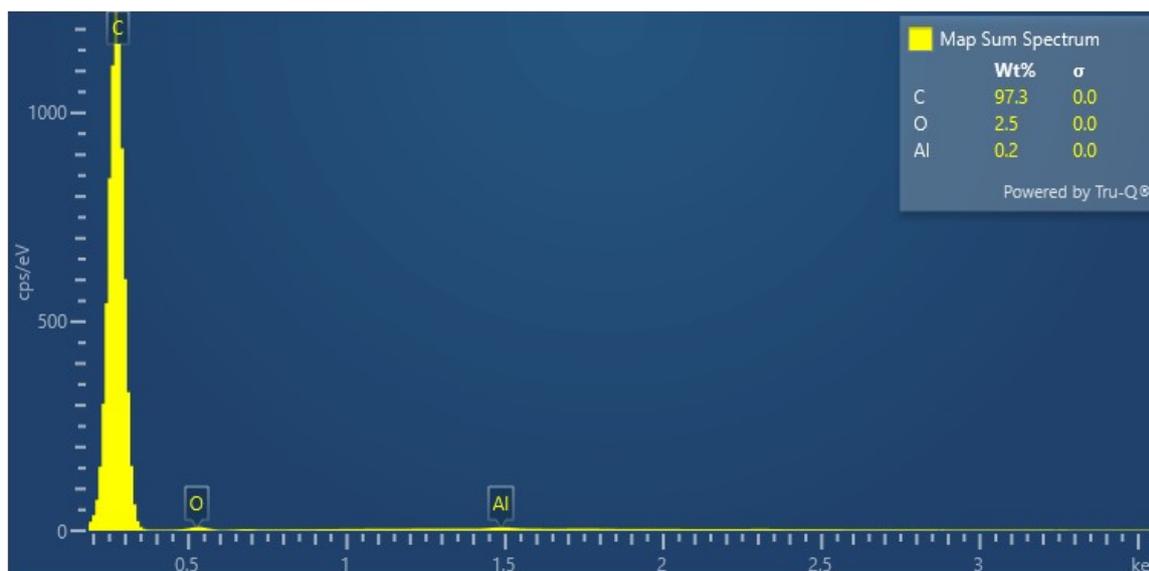**Fig. S1** Elemental mapping of commercial graphite.
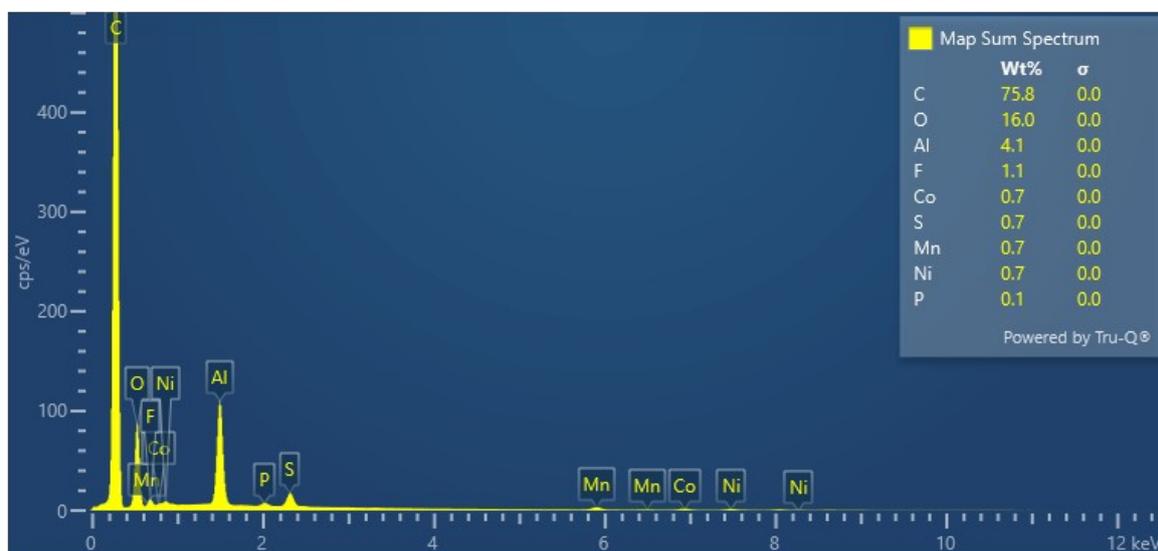
Fig. S2 EDS spectra and map sum spectrum of untreated SG.

Fig. S3 EDS spectra and map sum spectrum of commercial graphite.

Fig. S4 EDS spectra and map sum spectrum of sulfuric acid (2.75 mol/L, 240 min) purified SG.
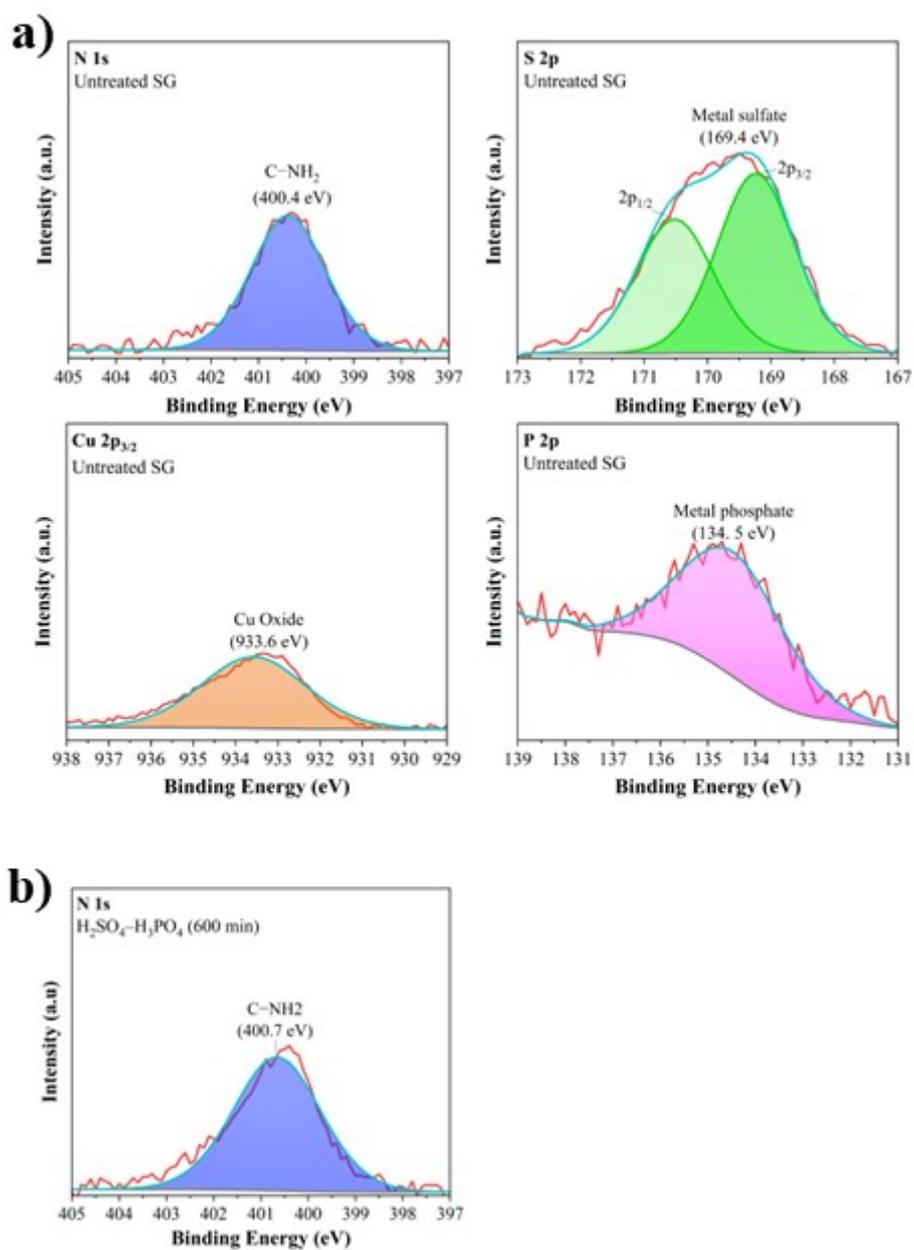
Fig. S5 EDS spectra and map sum spectrum of sulfuric acid–phosphoric acid (2.75 mol/L H_2SO_4-0.5 mol/L H_3PO_4, 600 min) purified SG.

Table S3. XPS results of untreated and H_2SO4–H_3PO_4 (600 min) treated SG (at. % = atomic %).

|	Chemical Bond	Untreated		H_2SO_4–H_3PO_4 (600 min)		Reference	
		BE (eV)	at. %	EV (eV)	at. %	BE (eV)	Source
Al 2p	Al Oxide	75.4	3.8	76.0	0.4	75.1–75.6	[1]
C 1s	C (Graphitic)	284.8	48.8	284.8	55.3	284.5–284.8	[2–4]
C 1s	C−C/C–H	285.9	4.2	285.8	4.3	284.8–285.9	[2–4]
C 1s	C−O	286.5	10.2	286.5	11.2	286–287	[2–4]
C 1s	C=O	288.8	4.9	288.7	7.5	288	[3]
C 1s	CF_2	291.1	3.3	291.1	5.7	290.0–291.0	[2–4]
F 1s	Organic F	688.0	3.6	688.0	3.9	687–688	[1,5]
N 1s	C−NH_2	400.4	0.6	400.7	0.8	400	A
P 2p	Metal phosphate	134.5	0.2	-	-	133	A
O 1s	Metal oxide	530.4	0.8	531.6	2.2	529	A
O 1s	Organic C−O	531.8	7.5	532.4	4.0	531.5–532	A
O 1s	Organic C=O	532.8	8.3	533.6	4.0	533	A
O 1s	O−H	534.0	2.7	534.9	0.7	533.5	[6]
S 2p	Metal sulfate	169.2	0.5	-	-	169	A
S 2p	Metal sulfate	170.5	0.4	-	-	169	A
Cu $2p_{3/2}$	Cu Oxide	933.6	0.2	-	-	1) 932.7 Cu(I) oxide 2) 933.1 Cu(II) oxide	A

A: Avantage Program. BE: binding energy

Fig. S6 High resolution N1s, S2p, Cu2p$_{3/2}$ and P2p spectra of a) untreated SG, and b) sulfuric acid–phosphoric acid (2.75 mol/L H$_2$SO$_4$ - 0.5 mol/L H$_3$PO$_4$, 600 min) purified SG.

Table S4. Residual impurities of sulfuric acid–phosphoric acid purified SG samples by ICP-OES (mg/g ± mg/g) after microwave-assisted acid digestion ($n = 2$) and elemental (CHNS(O)) analysis (wt% ± SD) of the samples by elemental analyzer ($n = 6$). Other factors: sulfuric acid concentration 2.75 mol/L, reaction temperature 100 °C, and L/S ratio 10 mL/g.

PO_4 (mol/L)	Time (min)		ICP-OES (mg/g) Al	Co	Cu	Li	Mn	Ni	P	Elemental analyzer (wt%) N	C	H	S	O
0.5	360	Av.	15.81	0.088	0.033	0.075	0.086	0.092	0.590	0.0	90.8	0.3	0	1.8
		±	1.06	0.016	0.001	0.003	0.016	0.016	0.008	0.1	0.2	0.0	0	0.0
0.5	480	Av.	8.37	0.074	0.029	0.051	0.070	0.086	0.618	0.1	92.7	0.3	0	1.7
		±	0.24	0.010	0.003	0.021	0.008	0.008	0.039	0.1	0.4	0.0	0	0.1
0.7	480	Av.	4.87	0.060	0.030	0.030	0.060	0.072	0.563	0.0	93.6	0.3	0.0	n.a
		±	0.21	0.004	0.000	0.000	0.003	0.003	0.002	0.0	0.3	0.0	0.1	n.a
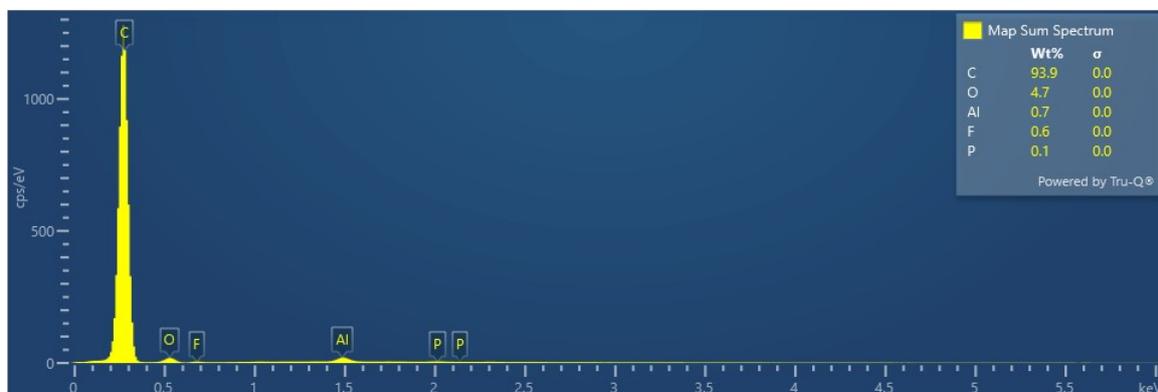
Fig. S7 EDS spectra and map sum spectrum of sulfuric acid–lithium phosphate (2.75 mol/L H_2SO_4 - 0.5 mol/L Li_3PO_4, 600 min) purified SG (Fig. 11a).

Fig. S8 EDS spectra and map sum spectrum of sulfuric acid–lithium phosphate (2.75 mol/L H_2SO_4 - 0.5 mol/L Li_3PO_4, 600 min) purified SG (Fig. 11b).

References

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