

Supporting Information

From Waste to Wealth: Upcycling Spent LiCoO₂ Batteries into a High-Performance Co₃O₄-Graphene Modified Diaphragm for Lithium-Sulfur Batteries

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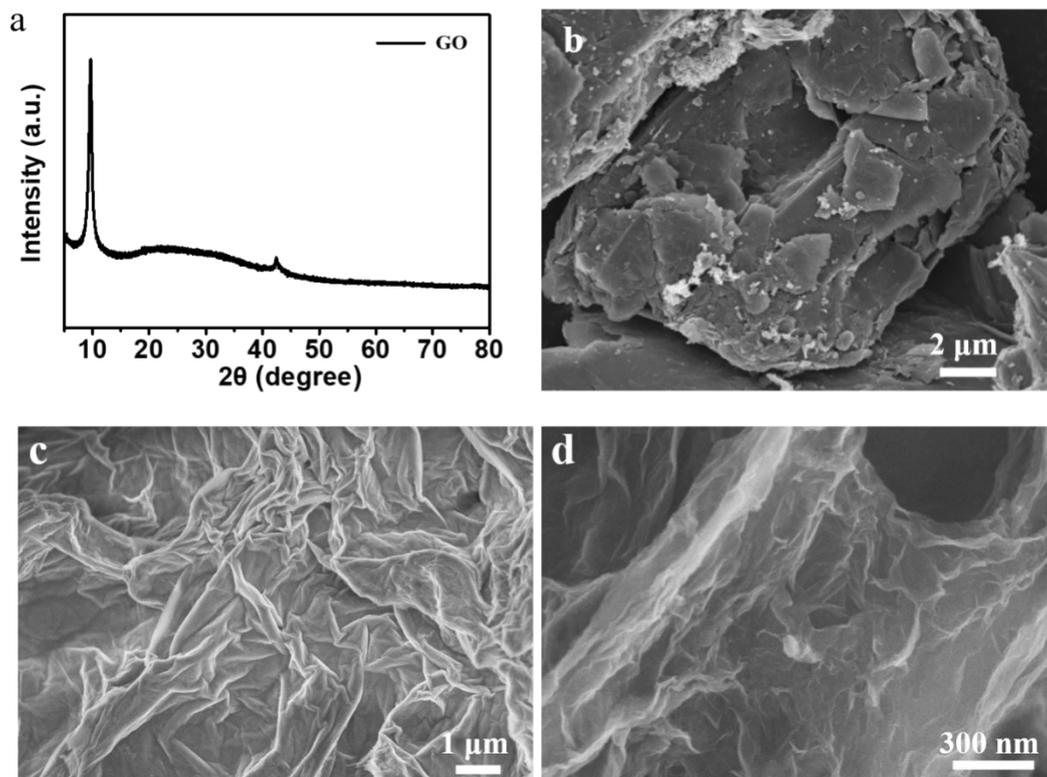


Fig. S1. (a) XRD pattern of GO. SEM images of (b) spent graphite powder, (c) GO and (d) rGO.

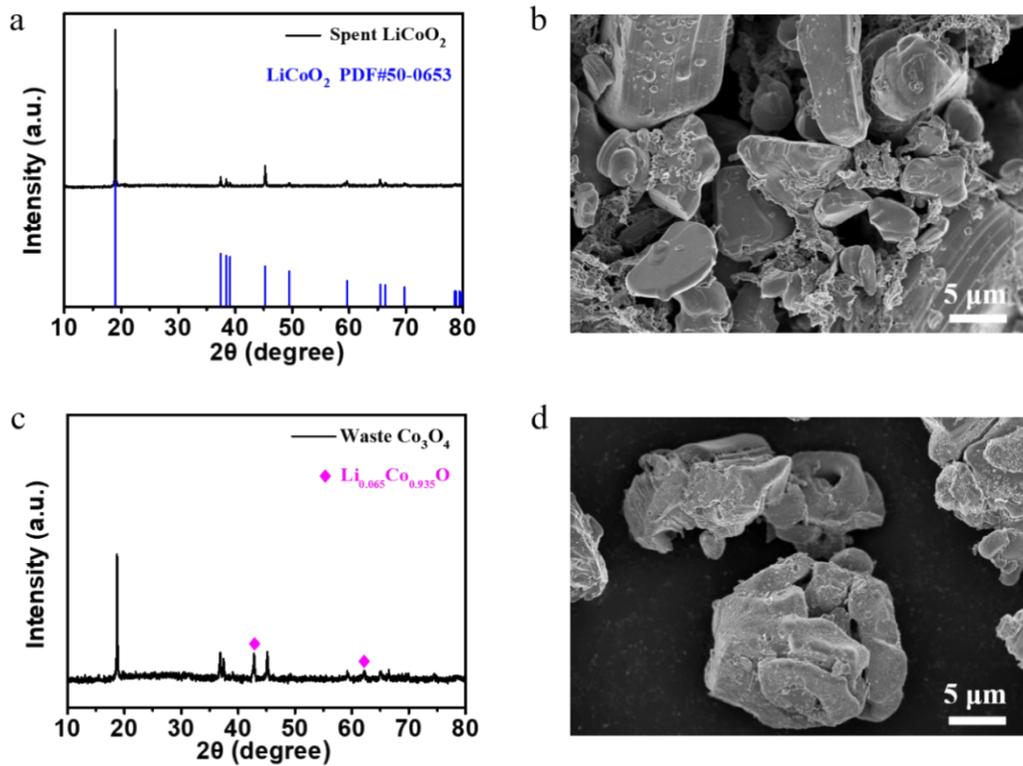


Fig. S2. (a) XRD pattern and (b) SEM image of spent LiCoO₂ powder; (c) XRD pattern and (d) SEM image of waste Co₃O₄ powder.

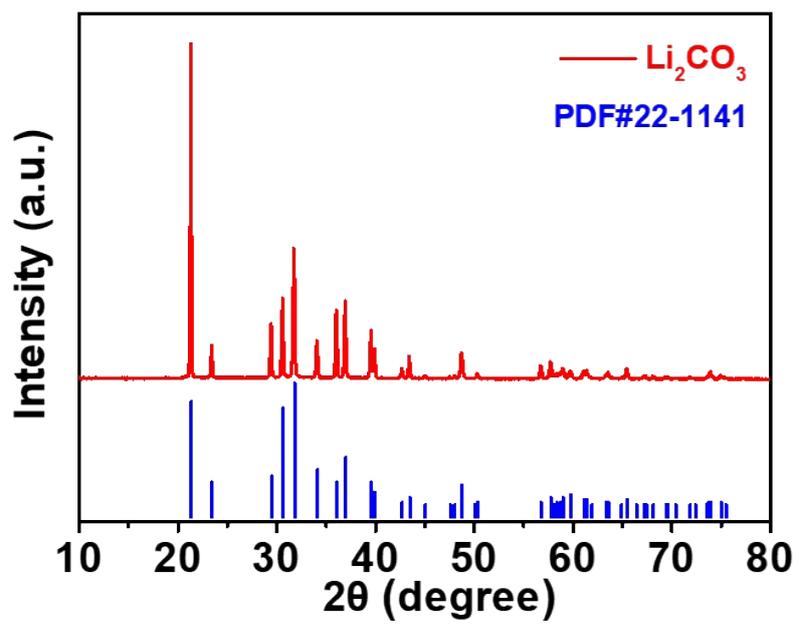


Fig. S3. XRD pattern of the as-prepared Li_2CO_3 powder.

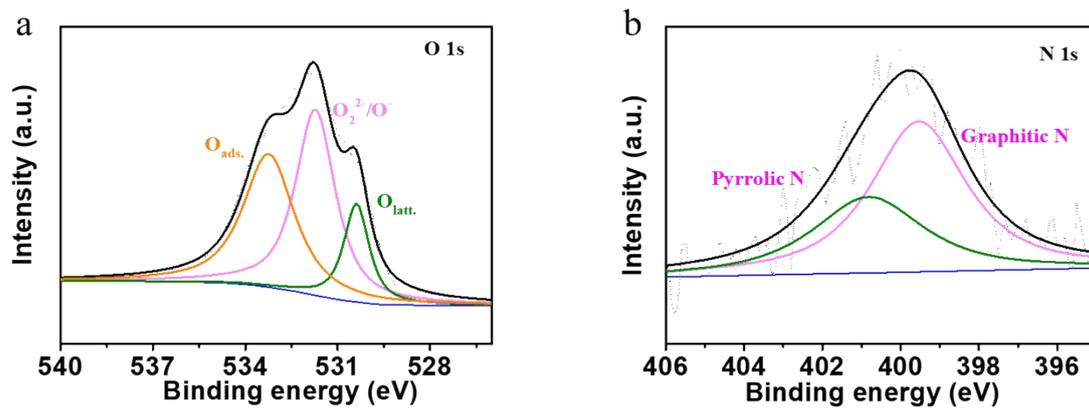


Fig. S4. High-resolution XPS spectra of (a) O 1s and (b) N 1s in Co_3O_4 -rGO-0.5.

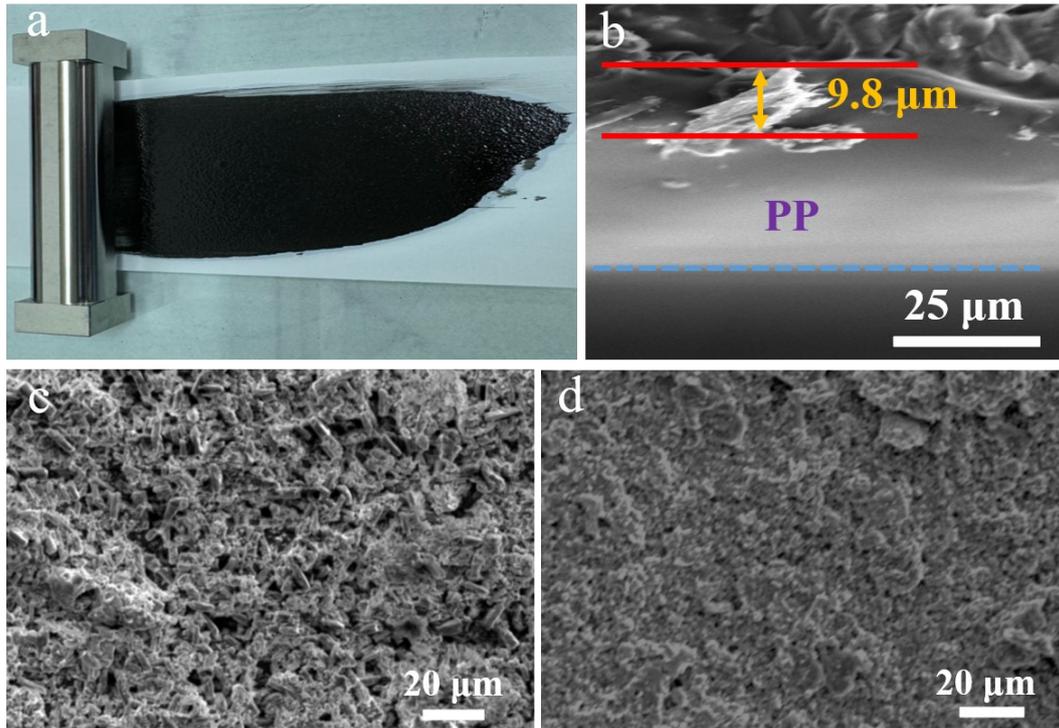


Fig. S5. (a) Photographs of homogeneous slurry containing $\text{Co}_3\text{O}_4\text{-rGO-0.5}$ doctor-bladed onto PP diaphragms. (b) Cross-sectional SEM image of $\text{Co}_3\text{O}_4\text{-rGO-0.5}$ modified diaphragms. SEM images of (c) WCo_3O_4 and (d) $\text{Co}_3\text{O}_4\text{-rGO-1.0}$ modified diaphragms.

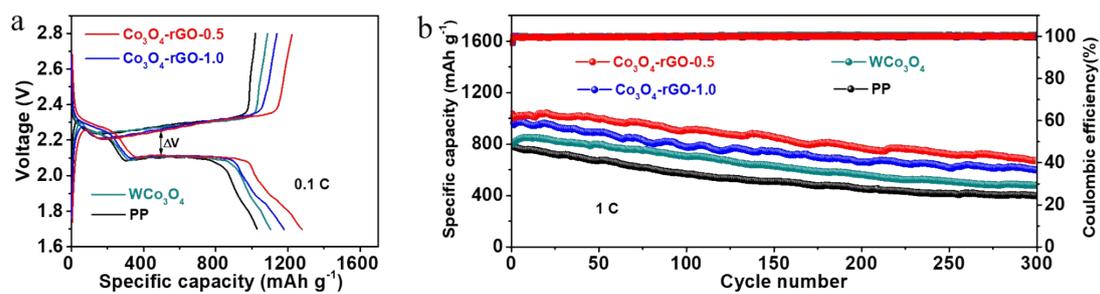


Fig. S6. (a) Galvanostatic discharge-charge profiles of Li-S batteries with bare PP, WCo₃O₄, Co₃O₄-rGO-0.5, and Co₃O₄-rGO-1.0 diaphragms at 0.1C. (b) Cycling performance of Li-S batteries with bare PP, WCo₃O₄, Co₃O₄-rGO-0.5, and Co₃O₄-rGO-1.0 diaphragms at 1C.

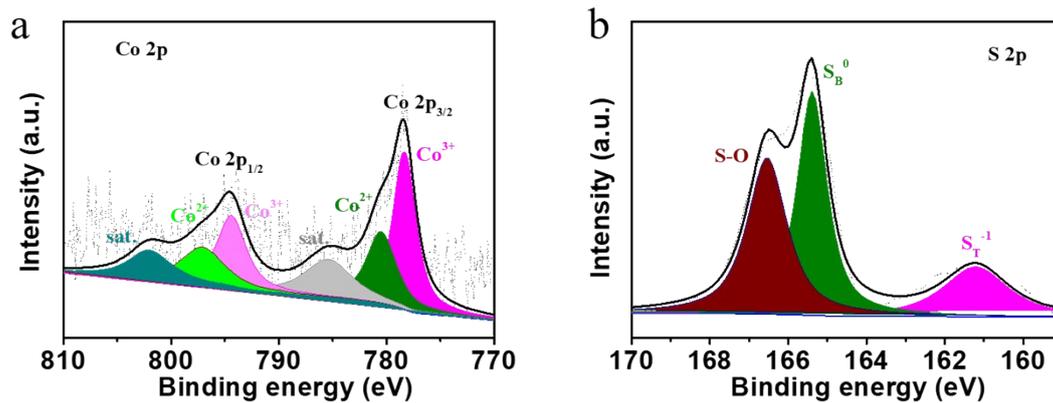


Fig. S7. High-resolution (a) Co 2p and (b) S 2p XPS spectra of the cycled Co₃O₄-rGO-0.5 modifier after 600 cycles at 3C.

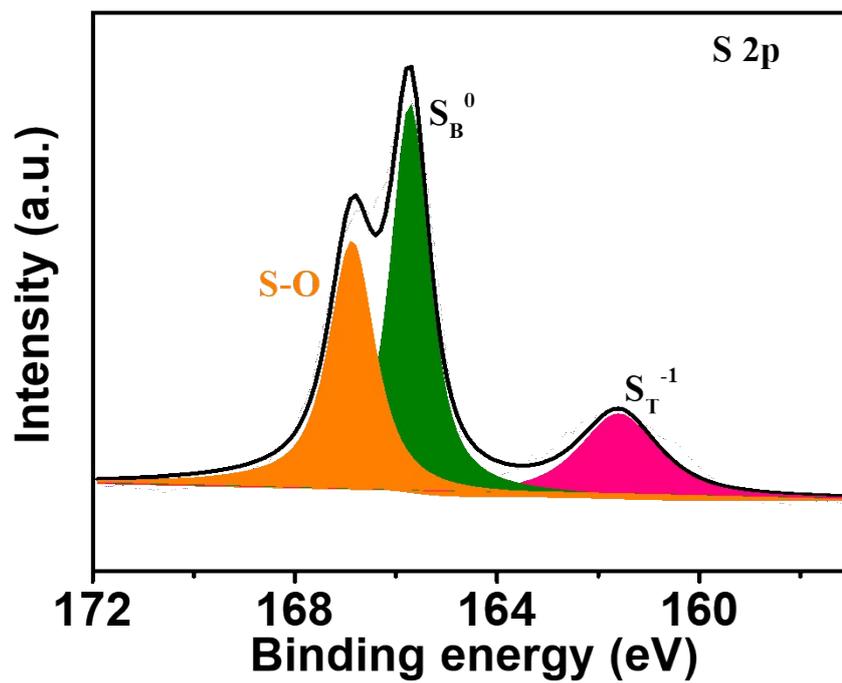


Fig. S8. High-resolution S 2p XPS spectra of Co_3O_4 -rGO-0.5 after immersing in Li_2S_6 solution.

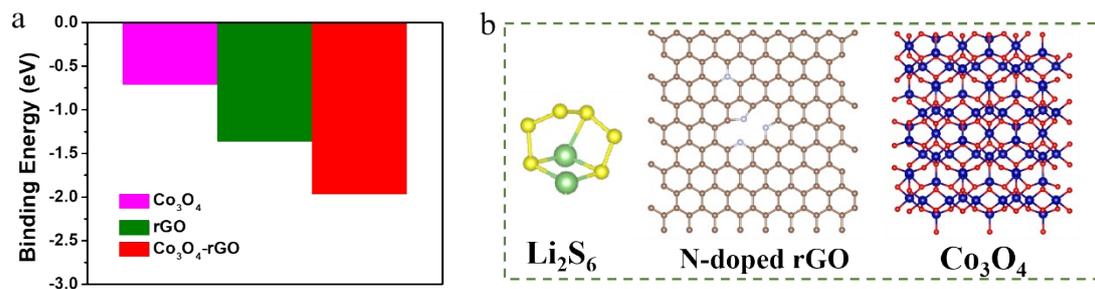


Fig. S9. (a) The summary of calculated binding energies between Li_2S_6 on Co_3O_4 (311), N-doped rGO, and Co_3O_4 -rGO; (b) Schematic configuration diagram of Li_2S_6 , N-doped rGO, and Co_3O_4 -rGO.

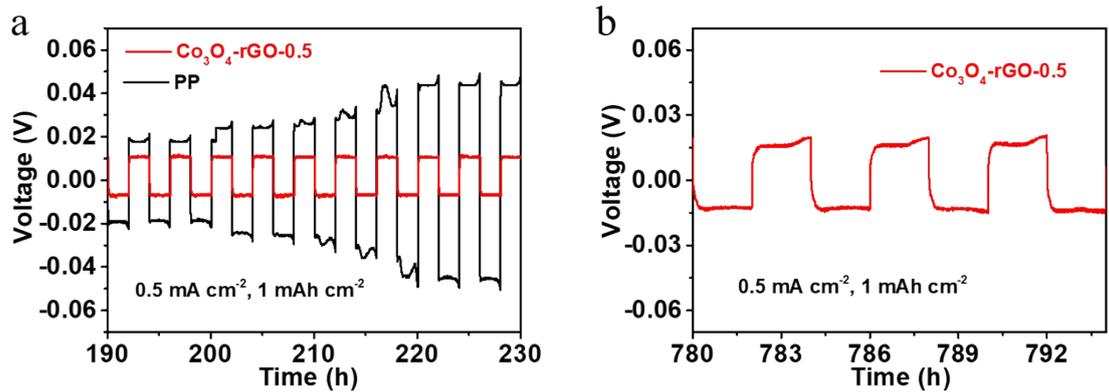


Fig. S10. The detailed voltage profiles of selected cycles of Li symmetric cells employing bare PP and $\text{Co}_3\text{O}_4\text{-rGO-0.5}$ modified diaphragms at 0.5 mA cm^{-2} with a fixed capacity of 1 mAh cm^{-2} .

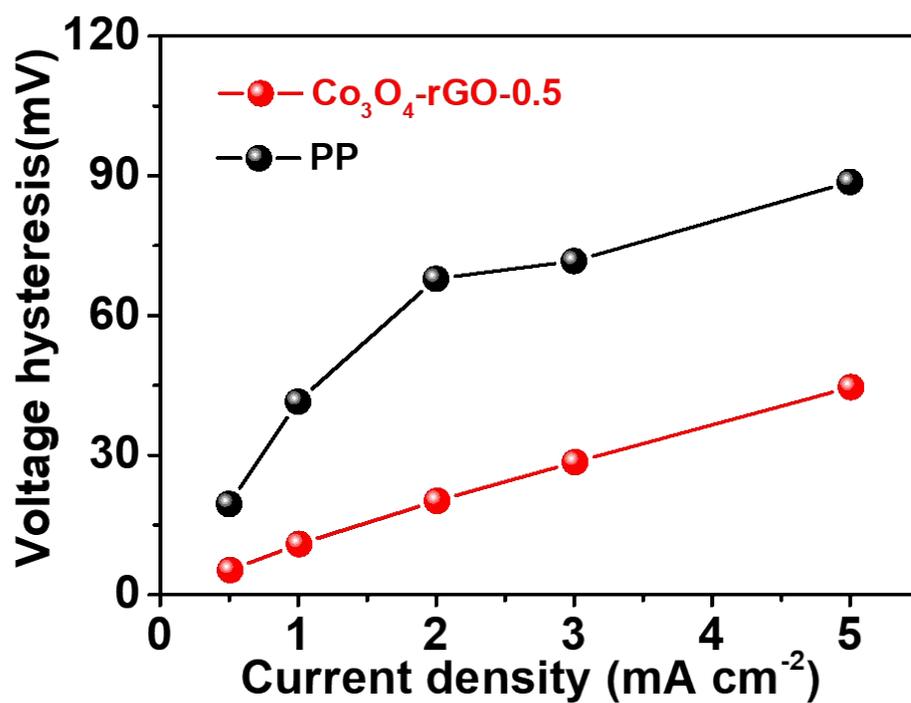


Fig. S11. Voltage hysteresis of Li symmetric cells employing bare PP and Co₃O₄-rGO-0.5 modified diaphragms at the increasing current densities (0.5 to 5 mA cm⁻²).

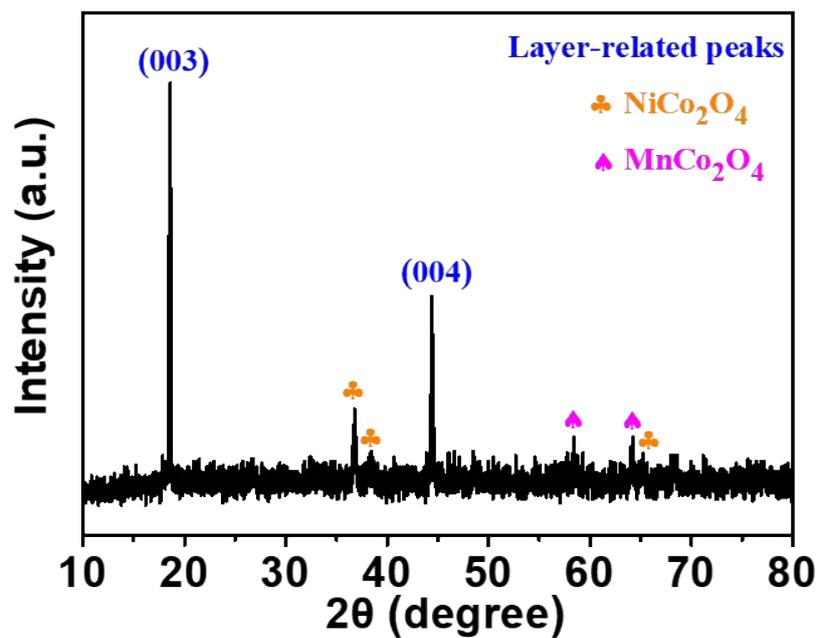


Fig. S12. XRD pattern of the composite derived from spent NMC (LiNi_{0.65}Mn_{0.28}Co_{0.07}O₂) cathode via the upcycling process.

Table S1. ICP-OES analysis of impurity elements in spent graphite and as-synthesized GO ($\times 10^{-4}$ mg/L).

Samples	Li	Al	Co	Cu	P
Spent graphite	375	311	98	862	394
Go	0	10	0	12	0

Table S2. Comparative life-cycle impact assessment: Virgin cobalt production vs. spent battery upcycling for cobalt oxide recovery.

Impact Category	Unit (per kg Co-eq)	Virgin cobalt production (Path A)	Spent battery upcycling (Path B)	Data sources
Global warming potential (GWP)	kg CO ₂ -eq	High (50~100+)	44.8	Ref. 48,49
Abiotic depletion potential (ADP)	kg Sb-eq	High	0.047	Ref. 48-50
Marine aquatic ecotoxicity potential (MAEP)	kg 1,4-DB eq	Very high	2.47×10^5	Ref. 48-50

Notes:

1. The qualitative descriptors (High, Very High) for Path A reflect the consensus in the life-cycle assessment literature on primary metal production, this is supported by typical database values (e.g., Ecoinvent) and aligns with the conclusion that its production is carbon-intensive (see, e.g., Ref. 47). The representative quantitative GWP value provided is a cradle-to-gate estimate for refined cobalt, included for illustrative comparison based on this reference.

2. All data for Path B are directly derived from our prior LCA study (Ref. 46), representing the sum of the “Pretreatment” and “Selective Leaching” stages to produce a cobalt oxide-rich residue. This provides a transparent, cradle-to-gate benchmark for our upcycling route.