Supplementary Information for

Co-upcycling spent lithium-ion batteries and plastics to microwave absorbing materials: Ni-Co catalyst size confinement and poison mitigation

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Text S1. Thermodynamic calculations

The standard Gibbs free energy, $\Delta_r G_T^\theta$, can be expressed as:

$$\Delta_r G = \Delta_r G_T^\theta + RT\ln(K_{P_B/P^\theta}) \quad (1)$$

where $\Delta_r G$ represents the Gibbs free energy of a certain reaction; R is the universal gas constant, 8.314 J mol$^{-1}\cdot$K^{-1}; T is the absolute temperature; K_{P_B/P^θ} represents gas pressure equilibrium constant. When a chemical reaction reaches its equilibrium state, $\Delta_r G = 0$ or $\Delta_r G_T^\theta = -RT\ln K$, while $\Delta_r G < 0$ indicates that the chemical reaction is spontaneous.

Thermal reduction involving C_2H_4:

$$12LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2 + C_2H_4(g) \rightarrow$$
$$12/5CoO + 36/5NiO + 12/5MnO + 6Li_2O + 2CO_2(g) + 2H_2O(g) \quad (2)$$

$$6NiO + C_2H_4(g) \rightarrow 6Ni + 2CO_2(g) + 2H_2O(g) \quad (3)$$

$$6CoO + C_2H_4(g) \rightarrow 6Co + 2CO_2(g) + 2H_2O(g) \quad (4)$$

$$6MnO + C_2H_4(g) \rightarrow 6Mn + 2CO_2(g) + 2H_2O(g) \quad (5)$$

Thermal reduction involving H_2:

$$2LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2 + H_2(g) \rightarrow 2/5CoO + 6/5NiO + MnO + Li_2O + H_2O(g) \quad (6)$$

$$NiO + H_2(g) \rightarrow Ni + 2H_2O(g) \quad (7)$$

$$CoO + H_2(g) \rightarrow Co + H_2O(g) \quad (8)$$

$$MnO + H_2(g) \rightarrow Mn + H_2O(g) \quad (9)$$

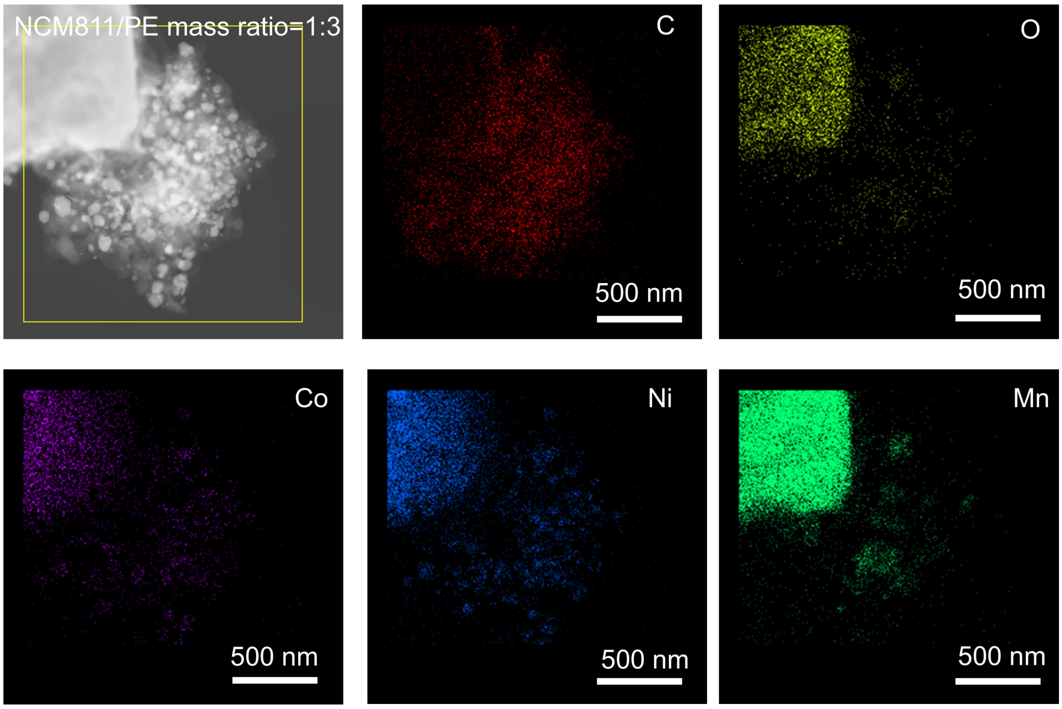
Text S2. Carbon conversion from binary plastics and MWCNT/NiCo mass ratio

$$\text{Carbon Conversion (\%)}=\left(\frac{C_{\text{CNT}}}{C_{\text{binary plastics}}}\right)\times 100 \tag{10}$$

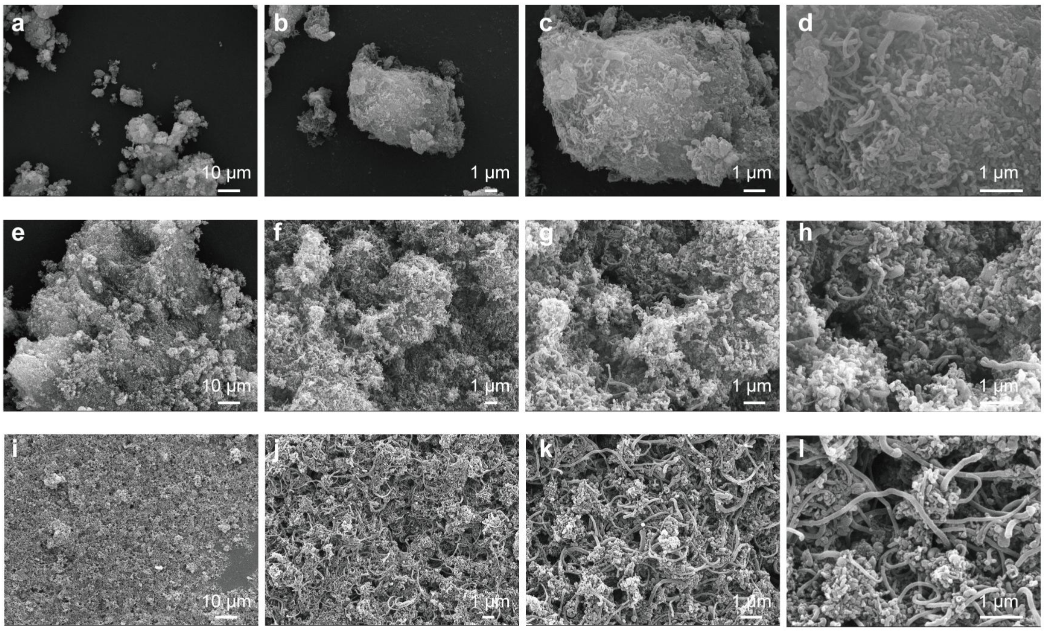
Where C_{CNT} is amount of carbon in the final CNT product, $C_{\text{binary plastics}}$ is amount of carbon in the binary plastics.

$$\text{MWCNT/NiCo mass ratio (\%)}=\frac{m_{\text{products}}-m_{\text{NiCoMnO in NCM}}}{m_{\text{NiCoMnO in NCM}}}\times 100 \tag{11}$$

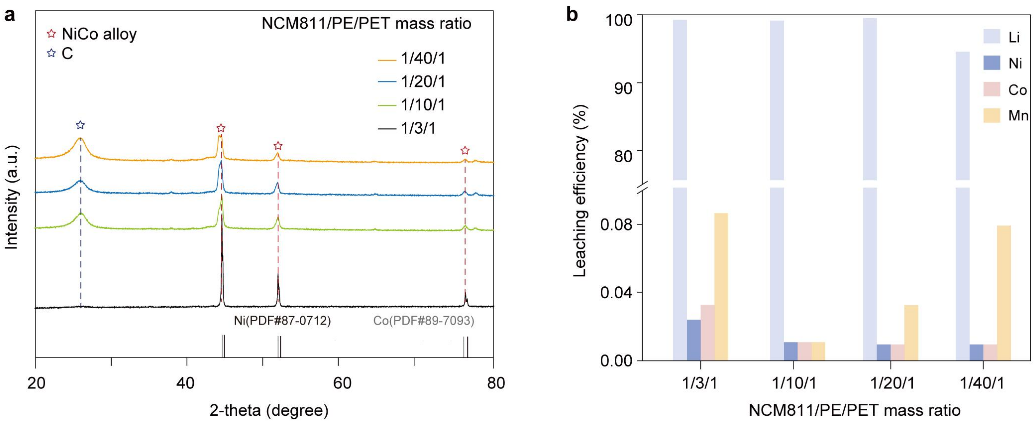
Where m_{products} is mass of products after water leaching, $m_{\text{NiCoMnO in NCM}}$ is the mass of NiCoMnO in the NCM.



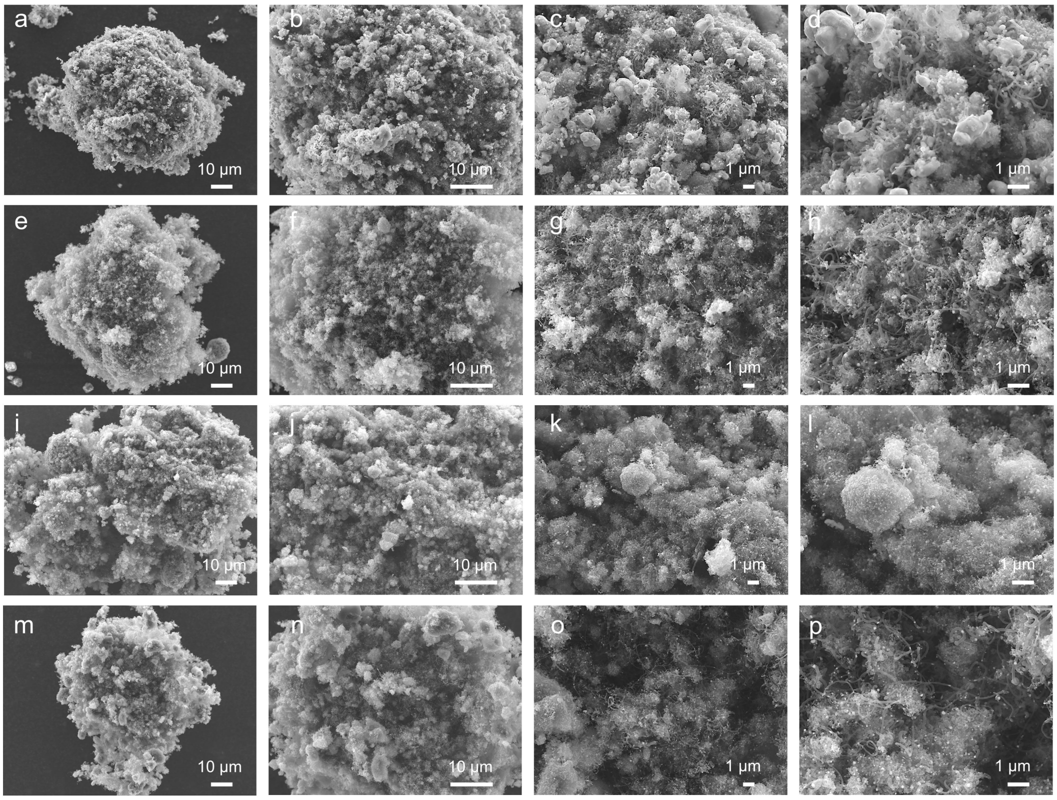
Supplementary Fig. 1. TEM and the corresponding elemental mapping images of NiCoMn@C.



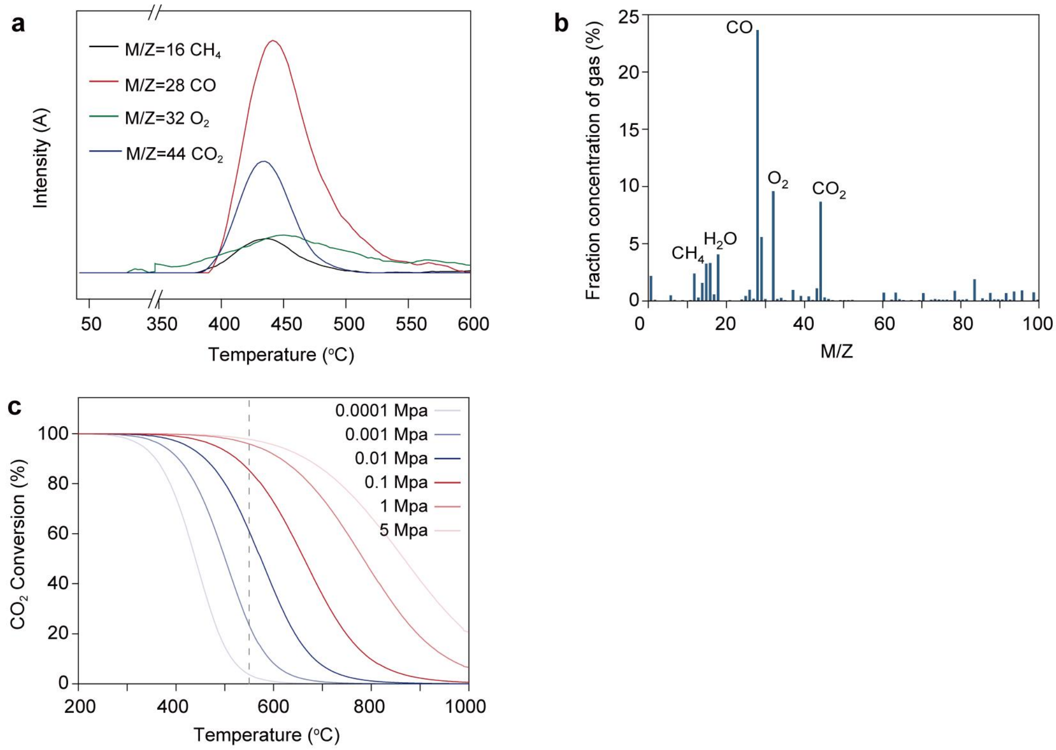
Supplementary Fig. 2. SEM images of products upcycled from spent NCM811 after co-pyrolysis reduction under various NCM811/PE/PET mass ratios (T = 550 °C, holding time = 5 h): **a-d,** 1:30:0, **e-h,** 1:30:0. 5, **i-l,** 1:30:1.



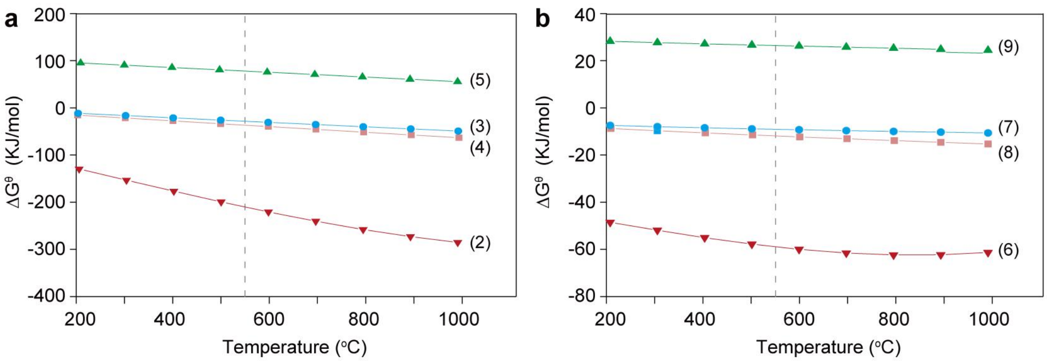
Supplementary Fig. 3. The products obtained using various NCM811/PE/PET mass ratios (holding time = 5 h, T = 500 °C), **a** XRD patterns and **b** Leaching efficiencies.



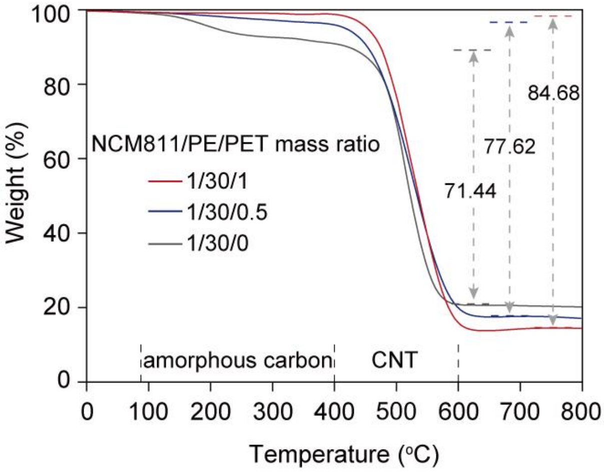
Supplementary Fig. 4. SEM images of products upcycled from spent NCM811 after co-pyrolysis reduction under various NCM811/PE/PET mass ratios (T = 550 °C, holding time = 5 h): **a-d,** 1:3:1, **e-h,** 1:10:1, **i-l,** 1:20:1, **m-p,** 1:40:1.



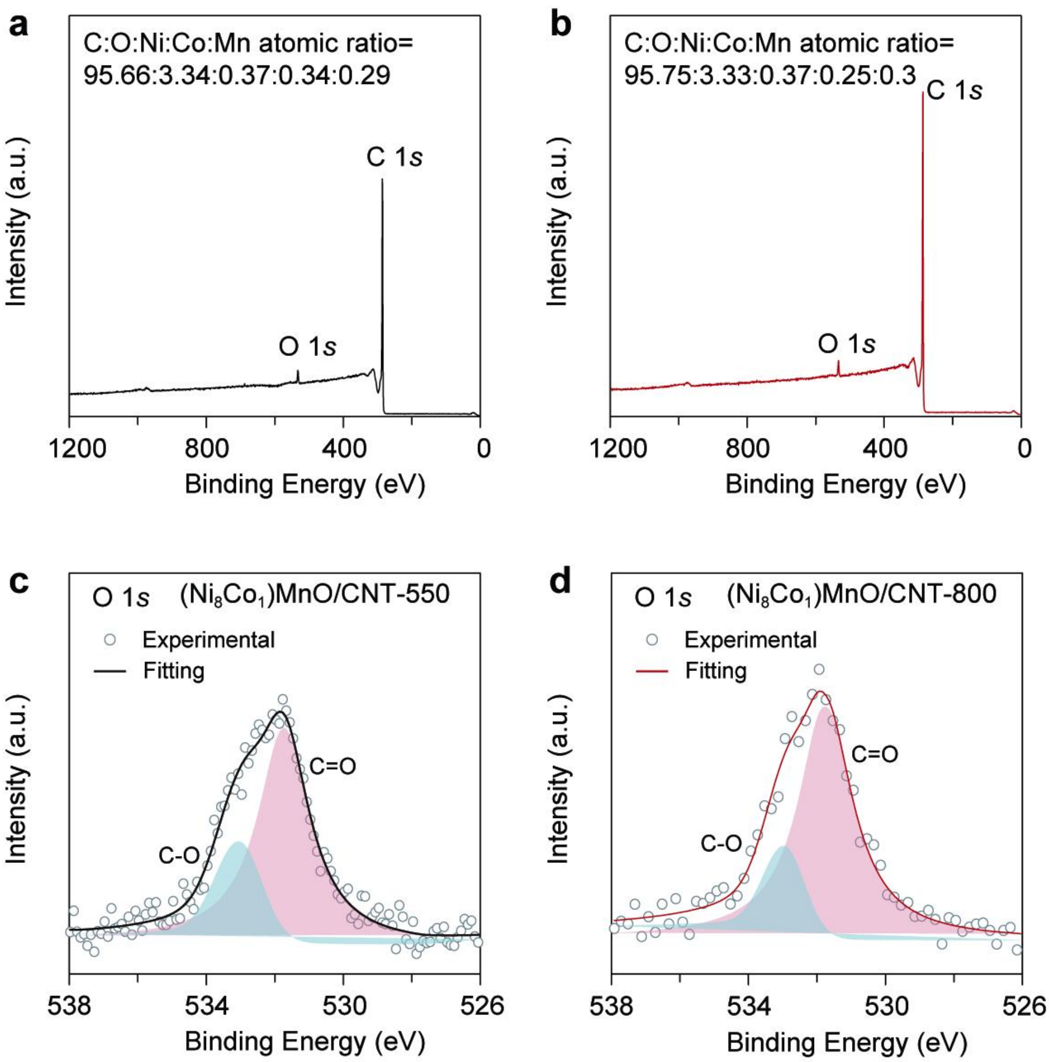
Supplementary Fig. 5. a, Mass spectrometric (MS) curves of polyethylene terephthalate (PET) measured in the range between 30 and 600 °C. **b,** Mass spectrum of PET pyrolysis gas. **c,** Relationship between the established gas composition and temperature for the gasification of CO_2.



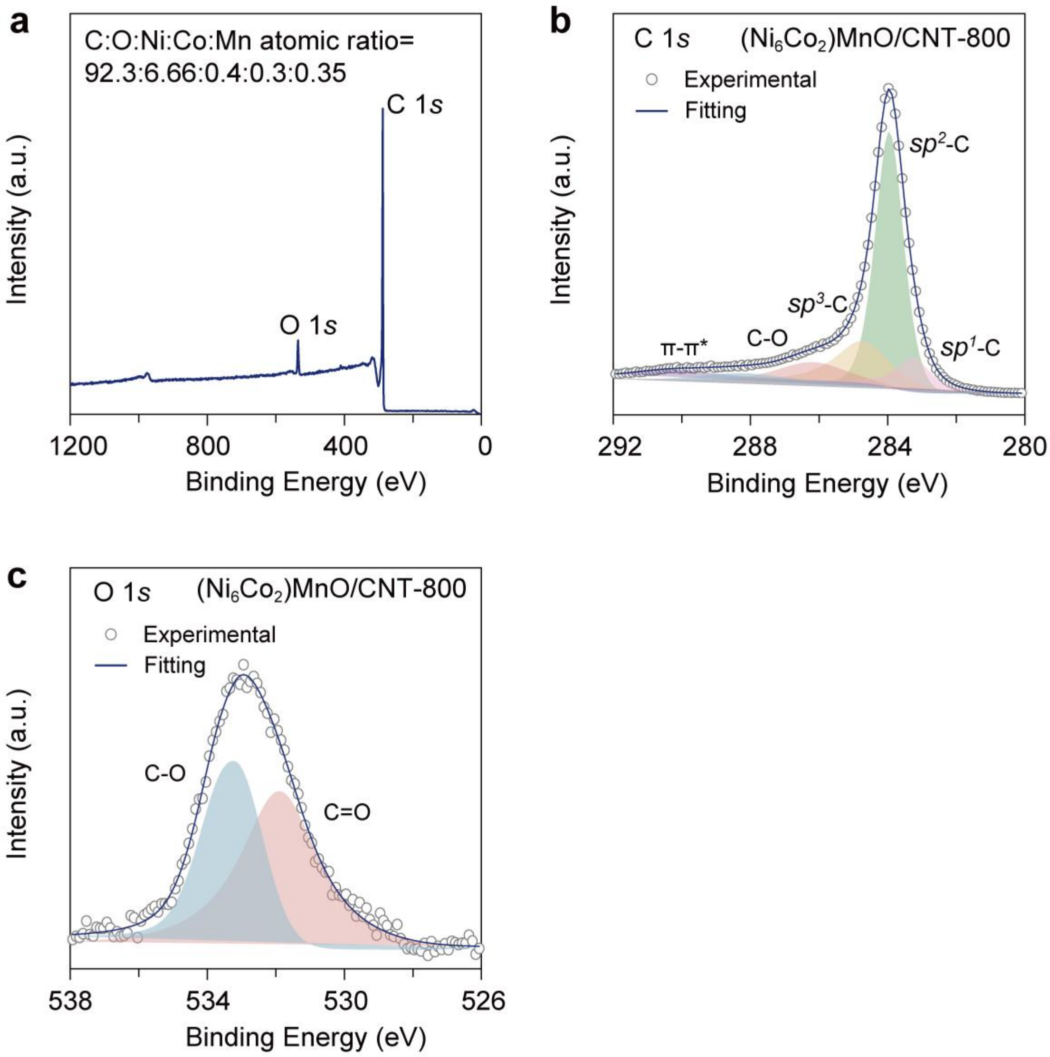
Supplementary Fig. 6. Elling diagrams illustrating the thermal reduction of cathode materials with **a,** C_2H_4 and **b,** H_2. Thermodynamic analysis was carried out using HSC 6.0 with the activity values of all involved species set to unity (a = 1).



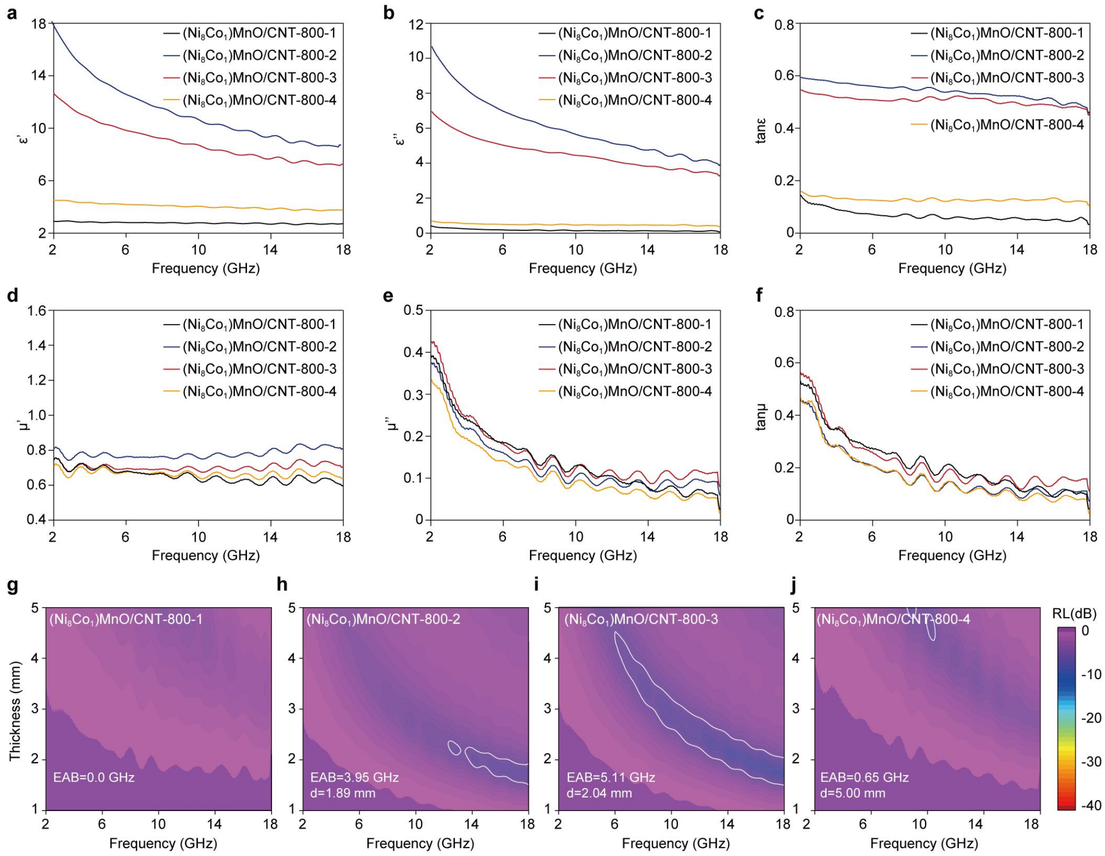
Supplementary Fig. 7. TG curves under air atmosphere of NCM811 products under various NCM811/PE/PET mass ratios. (550 °C, 5 h).



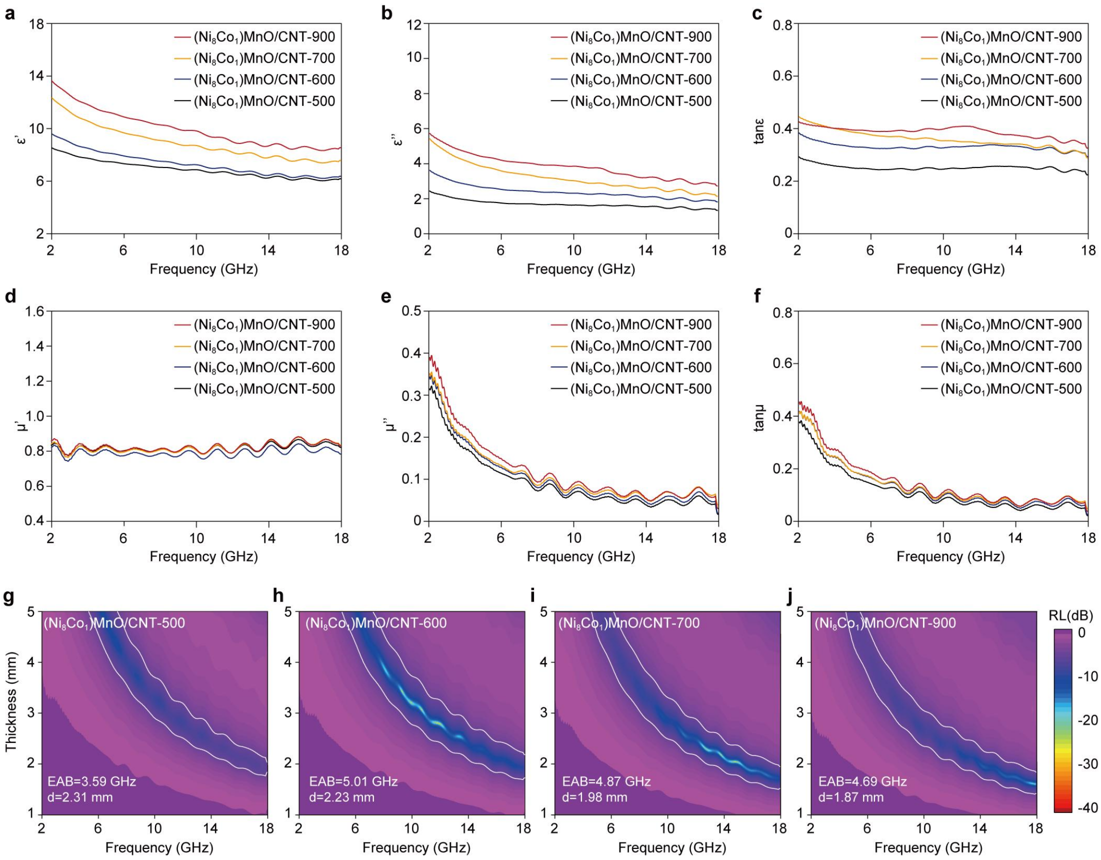
Supplementary Fig. 8. XPS full survey spectrum of **a,** $(Ni_8Co_1)MnO/CNT$ **b**, $(Ni_8Co_1)MnO/CNT$-800. **c**, O 1*s* $(Ni_8Co_1)MnO/CNT$-550. **d**, O 1*s* spectra of$(Ni_8Co_1)MnO/CNT$-800.



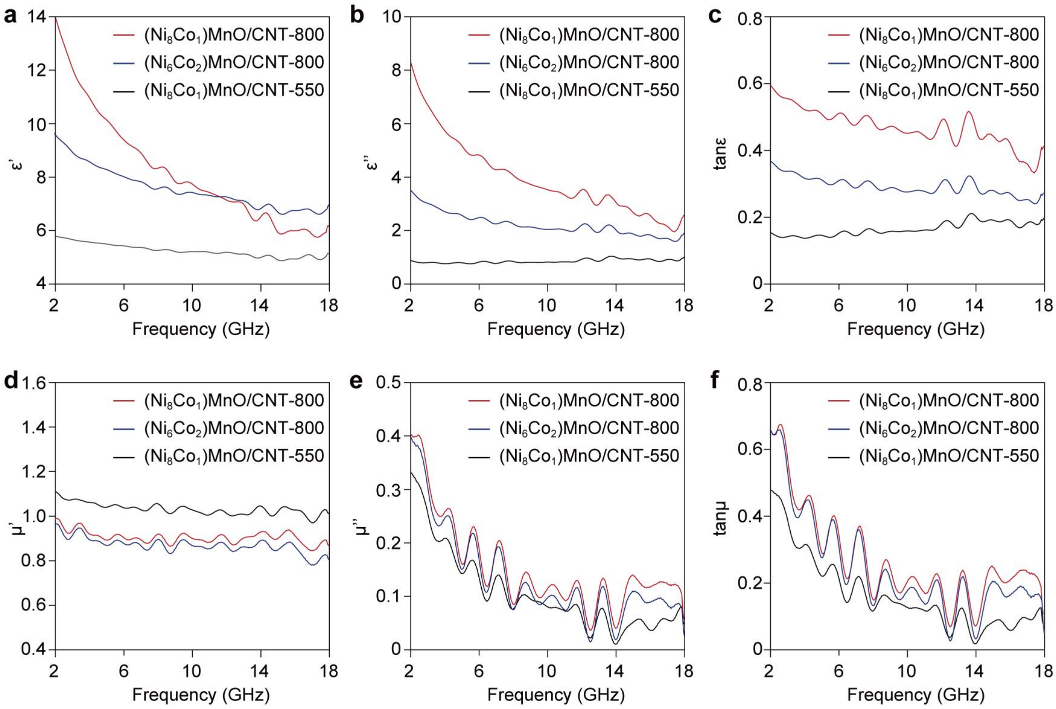
Supplementary Fig. 9. XPS full survey spectrum of **a,** $(Ni_6Co_2)MnO$/CNT-800. **b,** C 1*s*, and **c**, O 1*s* $(Ni_6Co_2)MnO$/CNT-800.



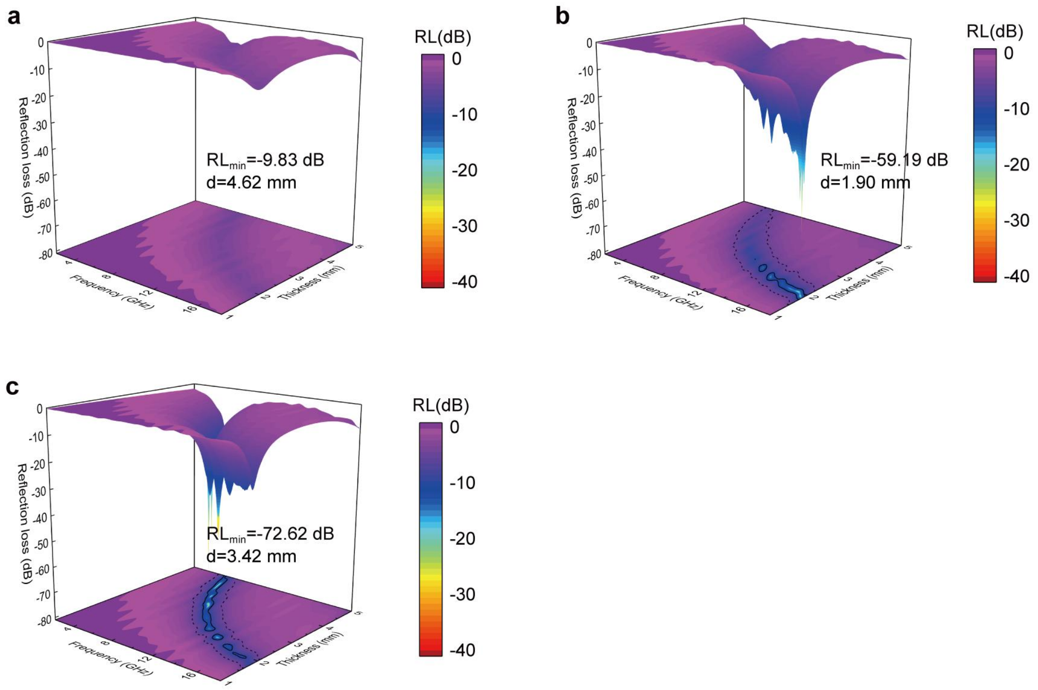
Supplementary Fig. 10. Dielectric properties and magnetic properties of (Ni$_8$Co$_1$)MnO/CNT-800-1, (Ni$_8$Co$_1$)MnO/CNT-800-2, (Ni$_8$Co$_1$)MnO/CNT-800-3 and, (Ni$_8$Co$_1$)MnO/CNT-800-4. **a,** real part ε' and **b,** imaginary part ε''. **c,** dielectric loss tangent, **d,** real part μ' and **e,** imaginary part μ'', and **f,** magnetic loss tangent; **g-h,** 2D RL value. The preparation conditions of the samples correspond to 550 °C, 5h, and the NCM811/PE/PET mass ratios are 1:3:1; 1:10:1; 1:20:1,1:40:1, respectively.



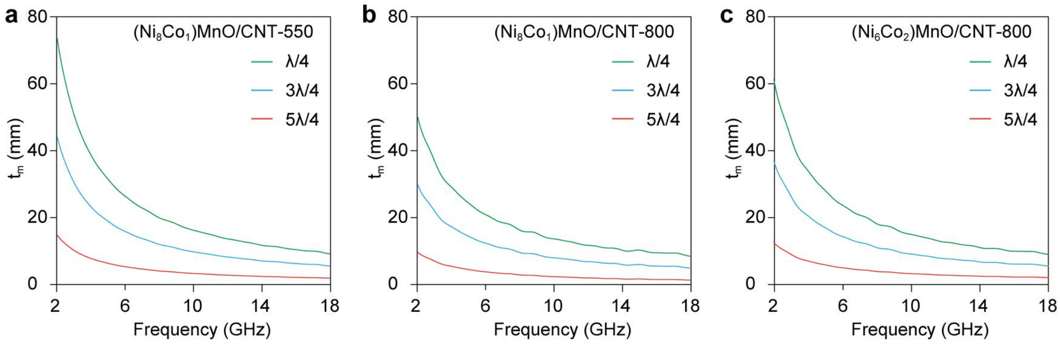
Supplementary Fig. 11. Dielectric properties and magnetic properties of $(Ni_8Co_1)MnO/CNT$-500, $(Ni_8Co_1)MnO/CNT$-600, $(Ni_8Co_1)MnO/CNT$-700 and, $(Ni_8Co_1)MnO/CNT$-900. **a,** real part ε' and **b,** imaginary part ε''. **c,** dielectric loss tangent, **d,** real part μ' and **e,** imaginary part μ'', and **f,** magnetic loss tangent; **g-h,** 2D RL value.



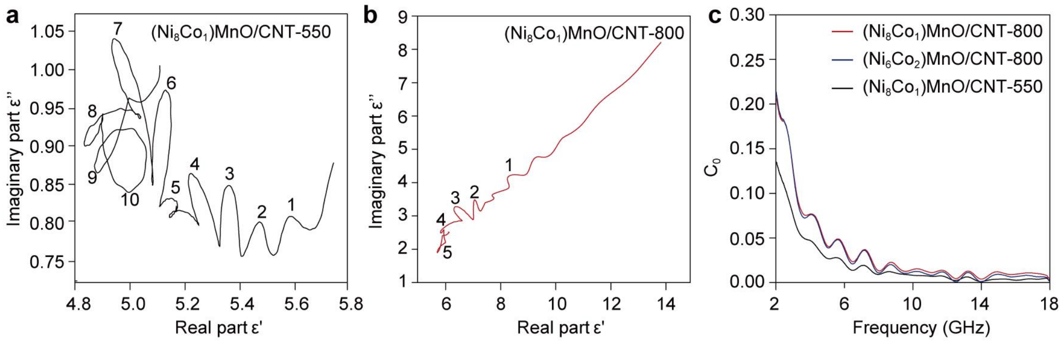
Supplementary Fig. 12. Dielectric properties and magnetic properties of $(Ni_8Co_1)MnO$/CNT-550, $(Ni_8Co_1)MnO$/CNT-800 and, $(Ni_6Co_2)MnO$/CNT-800. **a,** real part ε' and **b,** imaginary part ε''. **c,** dielectric loss tangent, **d,** real part μ' and **e,** imaginary part μ'', and **f,** magnetic loss tangent.



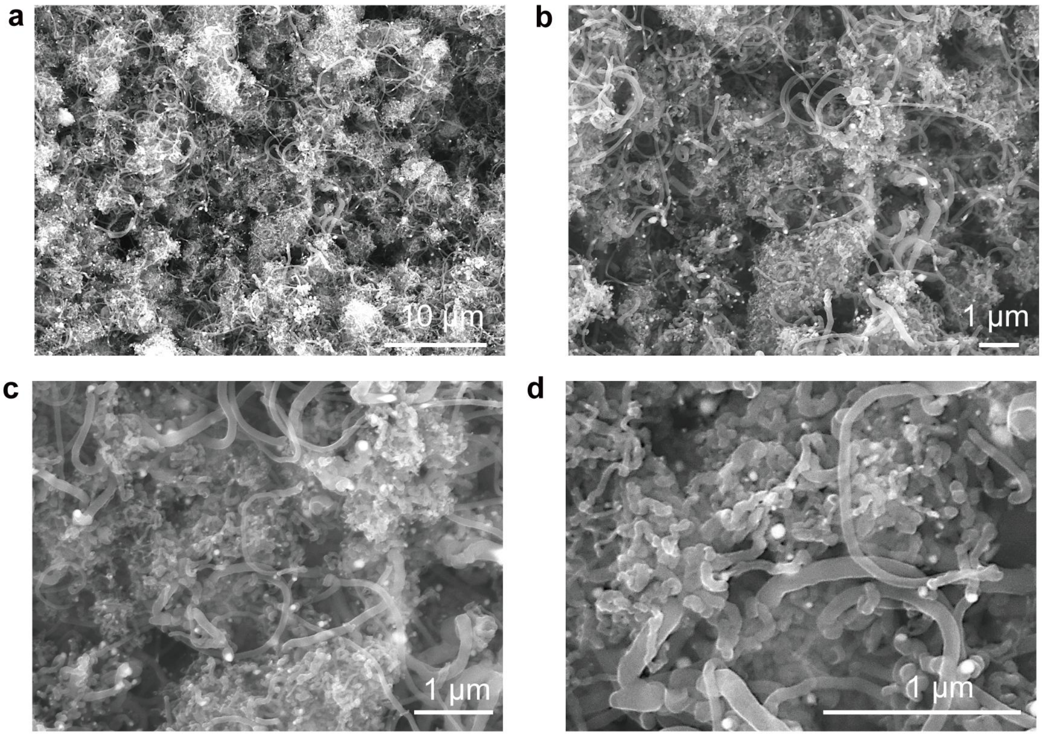
Supplementary Fig. 13. 3D RL values of **a,** $(Ni_8Co_1)MnO$/CNT-550, **b,** $(Ni_8Co_1)MnO$/CNT-800 and **c,** $(Ni_6Co_2)MnO$/CNT-800.



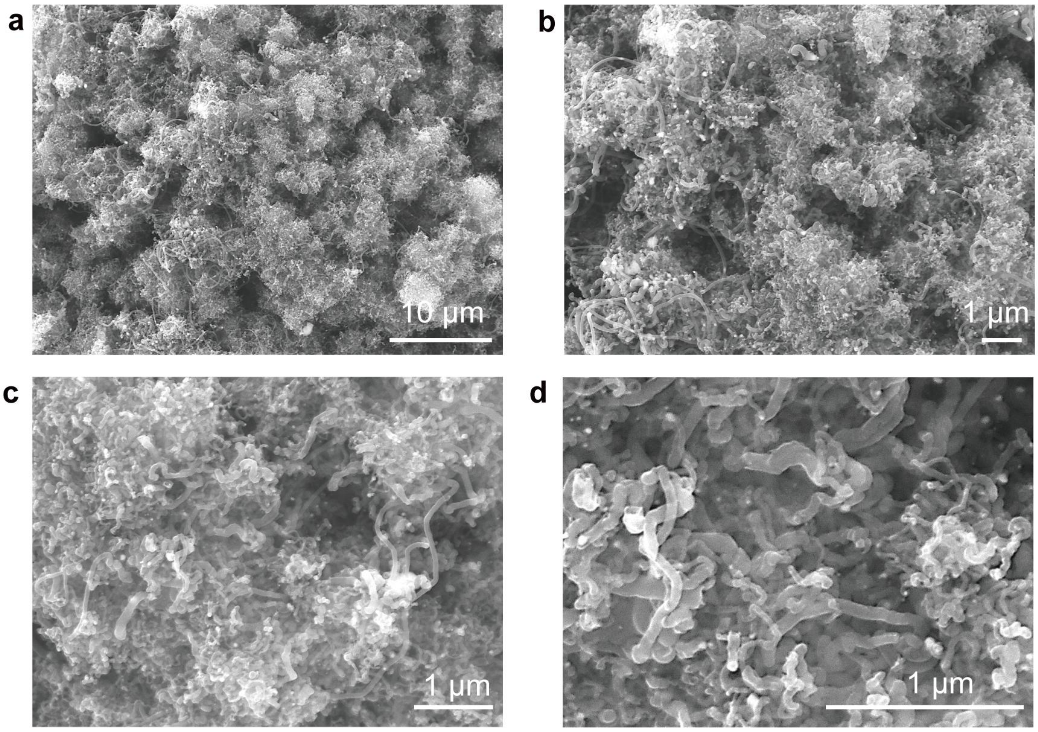
Supplementary Fig. 14. 1/4λ matching characteristics of **a,** $(Ni_8Co_1)MnO$/CNT-550, **b,** $(Ni_8Co_1)MnO$/CNT-800 and **c,** $(Ni_6Co_2)MnO$/CNT-800.



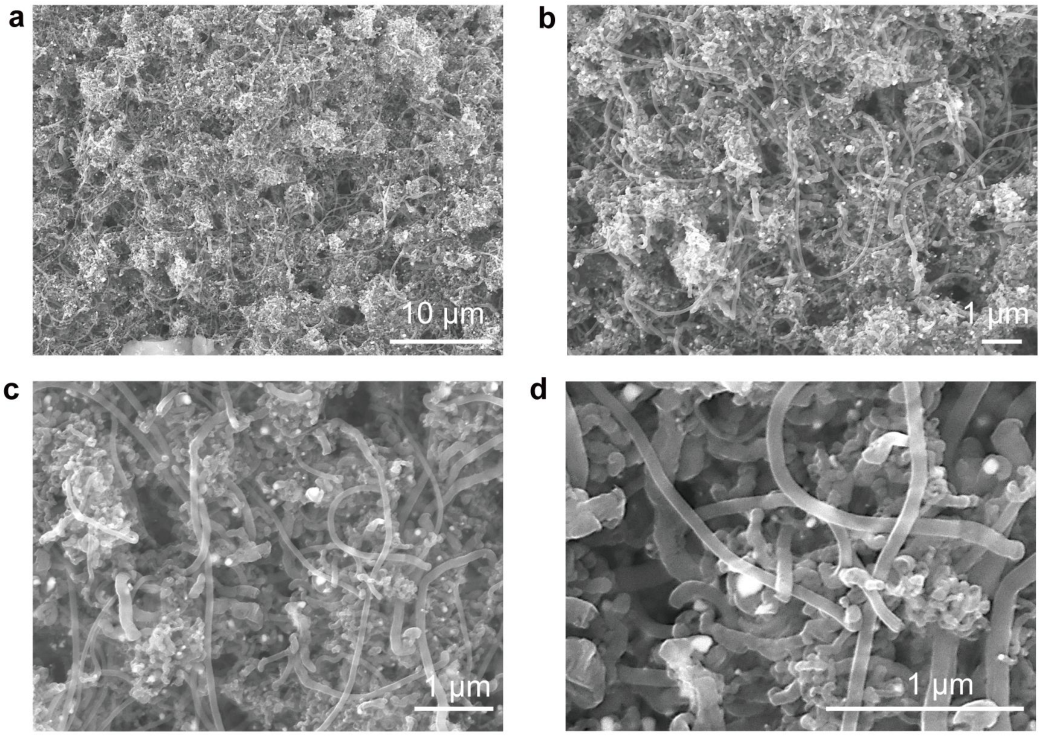
Supplementary Fig. 15. Cole-Cole curves of **a,** $(Ni_8Co_1)MnO$/CNT-550 and **b,** $(Ni_8Co_1)MnO$/CNT-800. **c,** eddy current loss of $(Ni_8Co_1)MnO$/CNT-550, $(Ni_8Co_1)MnO$/CNT-800 and, $(Ni_6Co_2)MnO$/CNT-800.



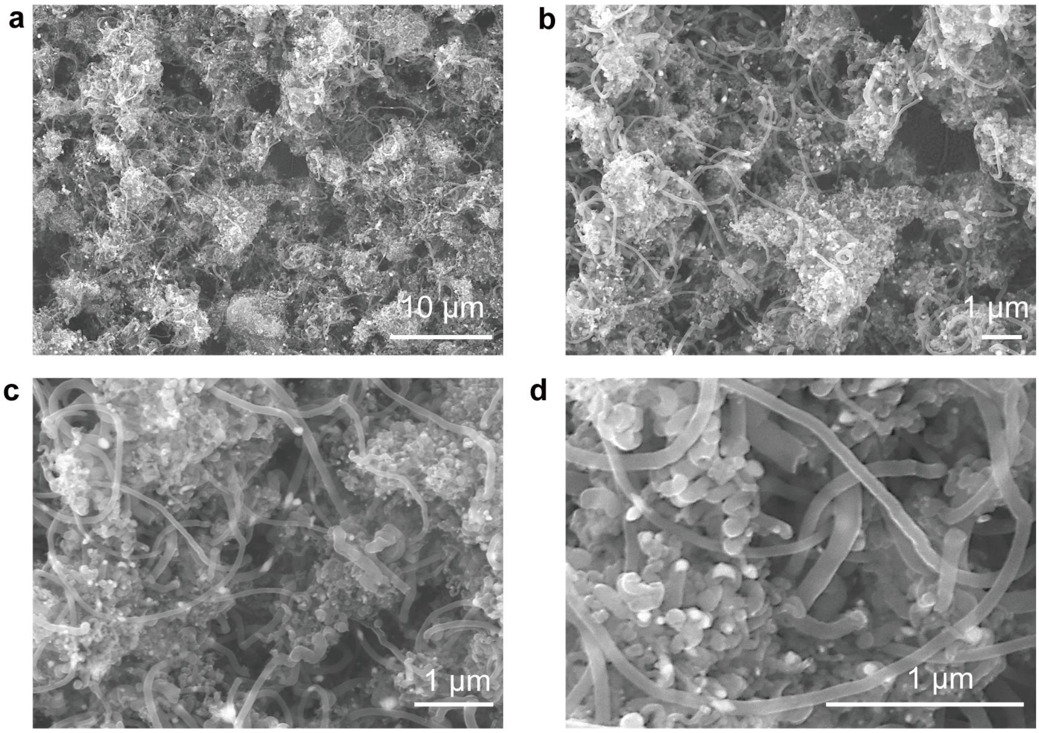
Supplementary Fig. 16. a-d, SEM images of products upcycled from spent NCM811 after co-pyrolysis reduction under NCM811 cathodes/LDPE/PET mass ratio = 1:30:1, 550 °C, 5 h.



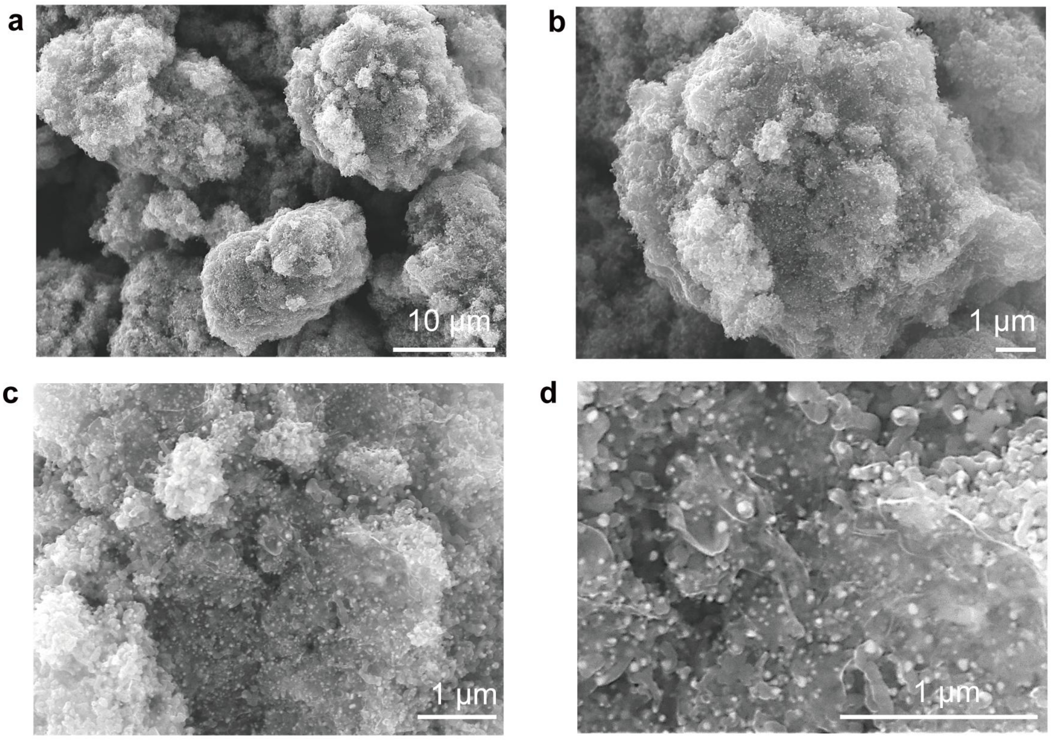
Supplementary Fig. 17. a-d, SEM images of products upcycled from spent NCM811 after co-pyrolysis reduction under of NCM811 cathodes/PP/PET mass ratio = 1:30:1, 550 °C, 5 h.



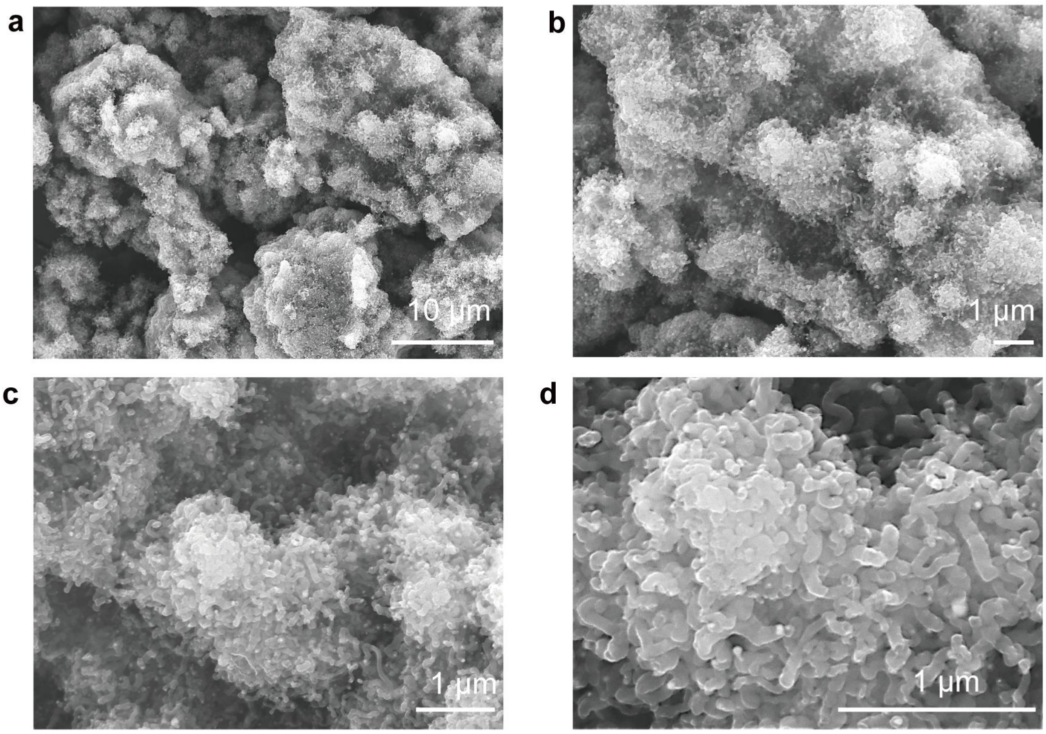
Supplementary Fig. 18. a-d, SEM images of products upcycled from spent NCM811 after co-pyrolysis reduction under of NCM811 cathodes/PC/PET mass ratio = 1:30:1, 550 °C, 5 h.



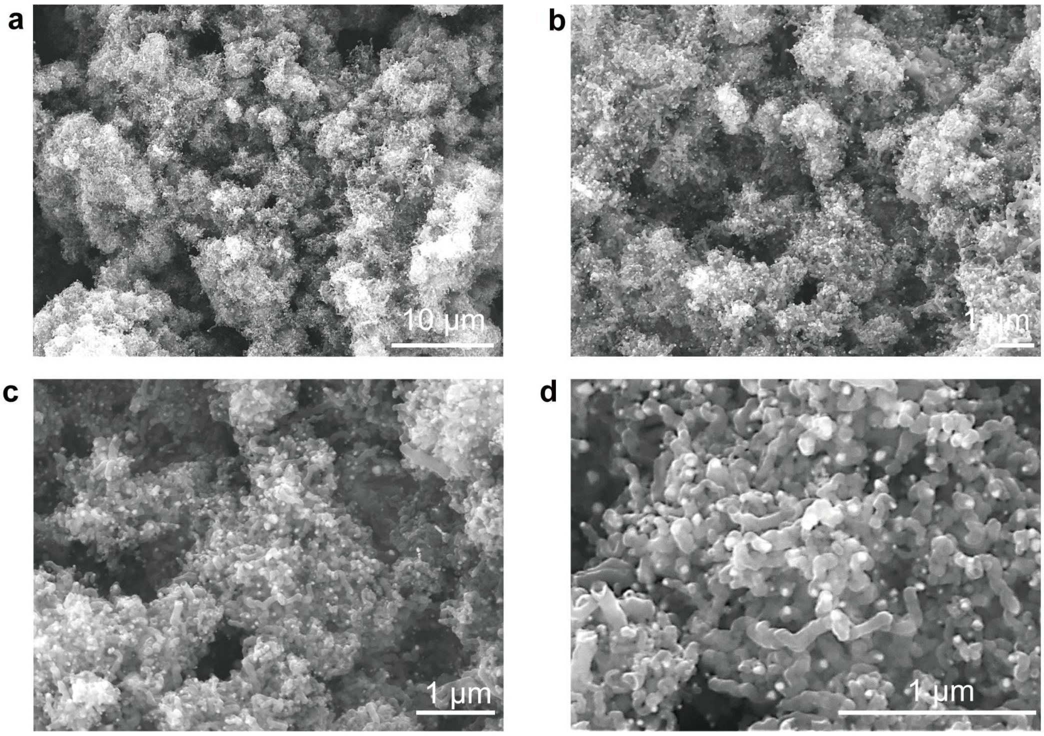
Supplementary Fig. 19. a-d, SEM images of products upcycled from spent NCM811 after co-pyrolysis reduction under of NCM811 cathodes/PS/PET mass ratio = 1:30:1, 550 °C, 5 h.



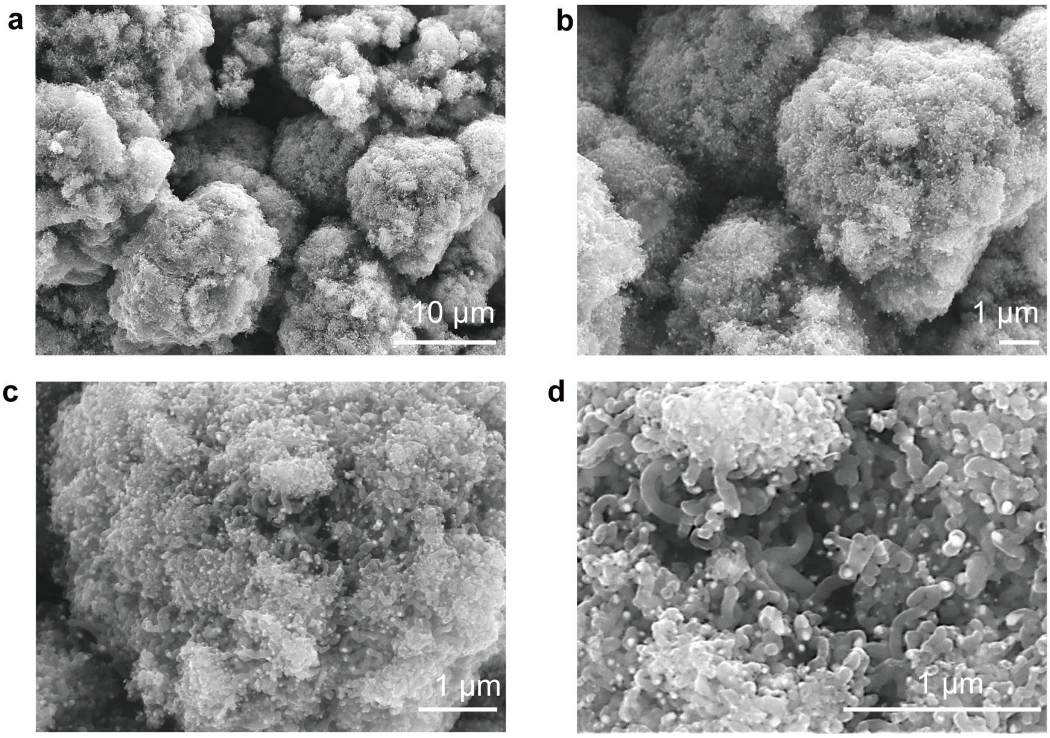
Supplementary Fig. 20. a-d, SEM images of products upcycled from spent NCM622 after co-pyrolysis reduction under of NCM622 cathodes/LDPE/PET mass ratio = 1:30:1, 550 °C, 5 h.



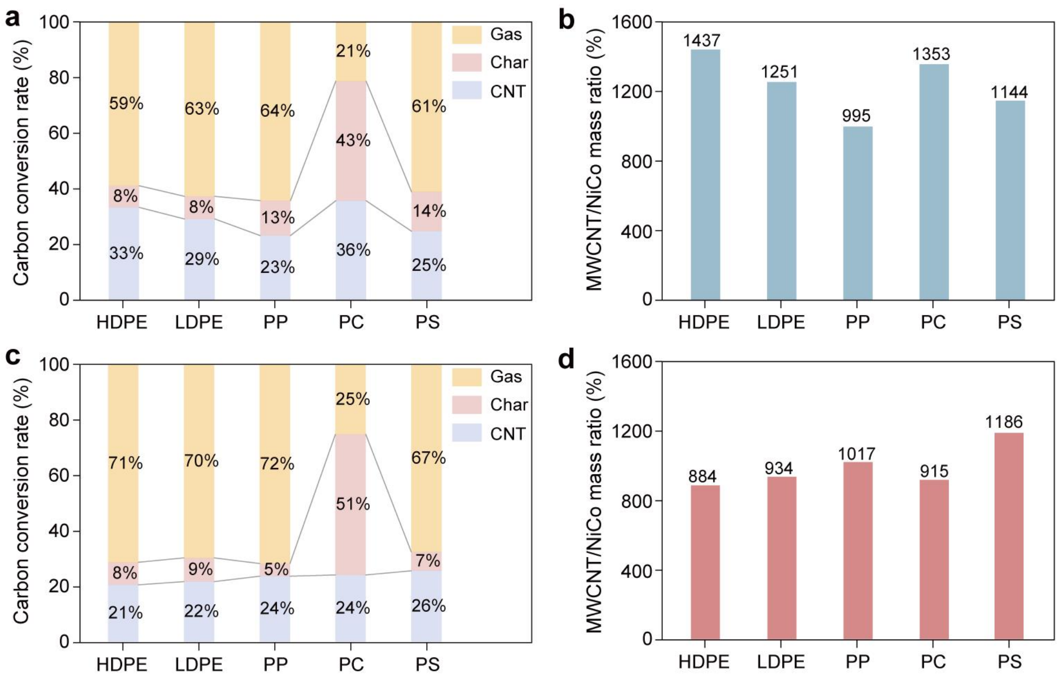
Supplementary Fig. 21. a-d, SEM images of products upcycled from spent NCM622 after co-pyrolysis reduction under of NCM622 cathodes/PP/PET mass ratio = 1:30:1, 550 °C, 5 h.



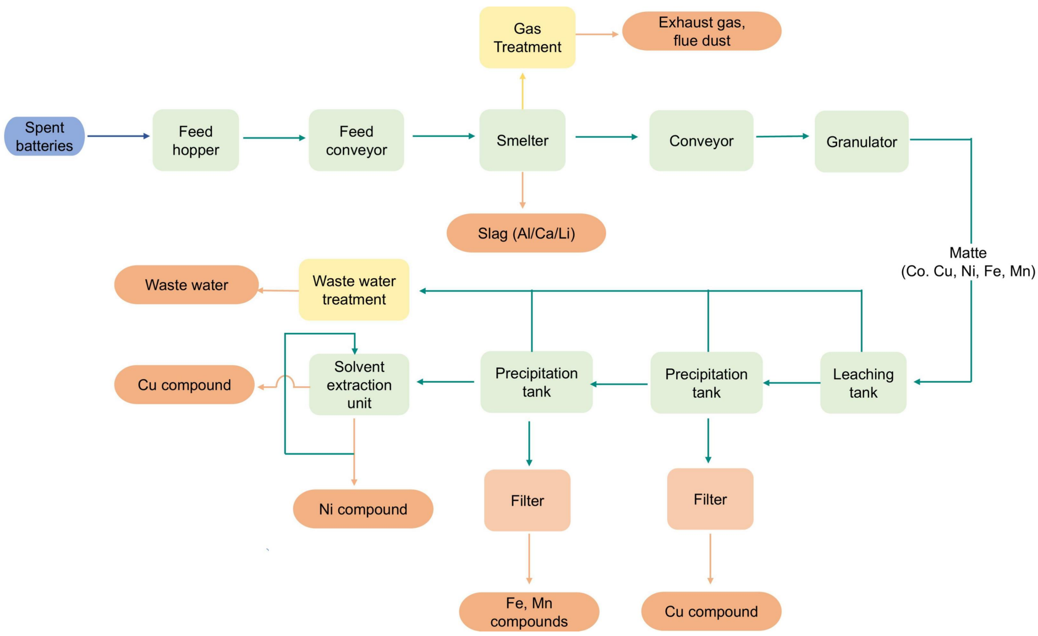
Supplementary Fig. 22. a-d, SEM images of products upcycled from spent NCM622 after co-pyrolysis reduction under of NCM622 cathodes/PC/PET mass ratio =1:30:1, 550 °C, 5 h.



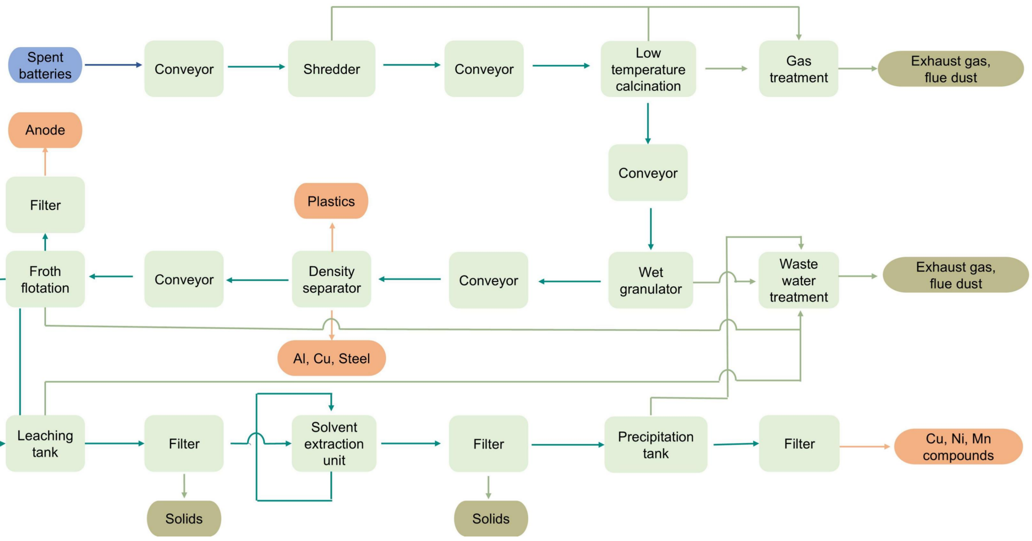
Supplementary Fig. 23. a-d, SEM images of products upcycled from spent NCM622 after co-pyrolysis reduction under of NCM622 cathodes/PS/PET mass ratio = 1:30:1, 550 °C, 5 h.



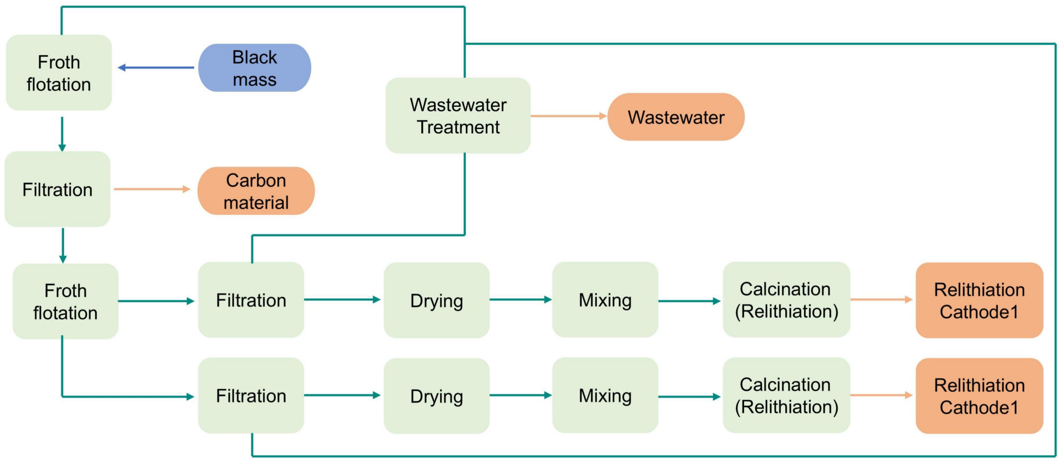
Supplementary Fig. 24. a, Carbon conversion rate of products upcycled from spent NCM811; **b,** MWCNT/NiCo mass ratio of products upcycled from spent NCM811; **c,** Carbon conversion rate of products upcycled from spent NCM622; **d,** MWCNT/NiCo mass ratio of products upcycled from spent NCM622; after co-pyrolysis reduction under of cathodes/plastics/PET mass ratio = 1:30:1, 550 °C, 5 h.



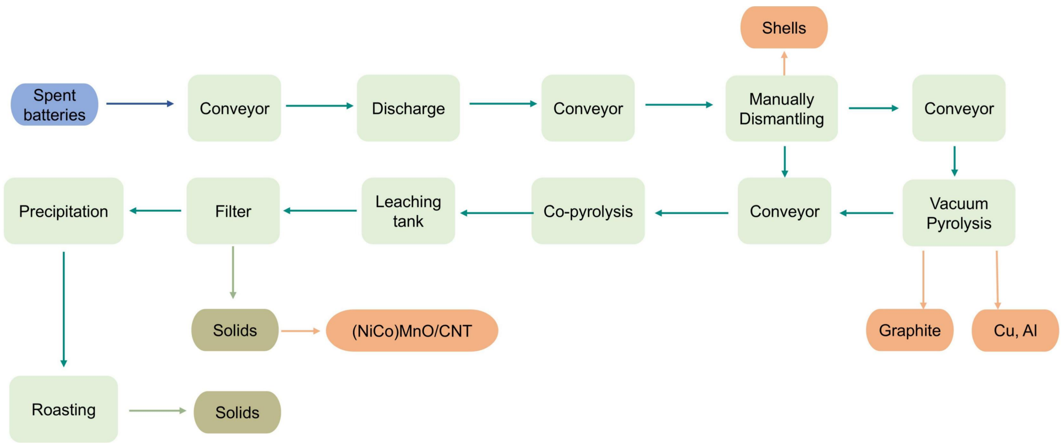
Supplementary Fig. 25. Flow chart of pyrometallurgy (Pyro) method.



Supplementary Fig. 26. Flow chart of hydrometallurgy (Hydro) method.



Supplementary Fig. 27. Flow chart of direct regeneration (Direct) method.



Supplementary Fig. 28. Flow chart of co-pyrolysis upcycling (Upcycling) method.

Supplementary Table 1. Comparative environmental impacts per 1 kg product (ReCiPe 2016 Midpoint (H))

Indicator	Unit	Pyrometallurgical	Hydrometallurgical	Direct	Upcycling
Global Warming Potential (GWP100)	kg CO_2 eq	0.71	0.36	0.34	0.10
Terrestrial Acidification Potential (TAP)	kg SO_2 eq	0.28	1.52	1.28	0.41
Fossil Resource Scarcity (FFP)	kg oil eq	0.74	2.77	2.93	0.86
Particulate Matter Formation (PMFP)	kg $PM_{2.5}$ eq	0.07	0.26	0.40	0.10
Water Consumption Potential (WCP)	m^3	0.05	0.18	0.25	0.06
Surplus Ore Potential (SOP)	kg Cu eq	1.31	3.15	2.19	0.45
Human Toxicity Potential (HTP, non-carcinogenic)	kg 1,4-DCB eq	0.02	0.04	0.04	0.02
Photochemical Oxidant Formation (HOPF)	kg NO_X eq	0.01	0.05	0.05	0.01

Supplementary Table 2. The environmental analysis of NCM811 recycling process

	Pyrometallurgical	**Hydrometallurgical**	**Direct**	**Upcycling**
Total Emissions (g/kg)				
VOC	0.71	0.36	0.34	0.10
CO	0.28	1.52	1.28	0.41
NO_x	0.74	2.77	2.93	0.86
PM10	0.07	0.26	0.40	0.10
PM2.5	0.05	0.18	0.25	0.06
SO_x	1.31	3.15	2.19	0.45
BC	0.02	0.04	0.04	0.02
OC	0.01	0.05	0.05	0.01
CH_4	0.56	4.87	3.89	1.19
N_2O	0.01	0.05	0.05	0.02
CO_2	1833	2261	2401	835
GHGs	1852	2423	2532	876

Supplementary Table 3. The economic analysis of NCM811 recycling process

	Pyrometallurgical	**Hydrometallurgical**	**Direct**	**Upcycling**
Revenue ($/kg)				
Copper metal	0.55	0	0	0
Co^{2+}	1.24	1.74	0	0
Ni^{2+}	3.31	4.64	0	0
Mn^{2+}	0	0.10	0	0
Li_2CO_3	0	1.69	0	0
Graphite	0	0.07	0.07	0.07
NCM811	0	0	14.04	0
$(Ni_8Co_1)MnO$/CNT	0	0	0	100
Cost ($/kg)				
Annualized capital cost	0.45	0.25	0.13	0.23
Other fixed costs	0.12	0.09	0.08	0.08
Plant overhead	0.08	0.13	0.18	0.17
maintenance	0.07	0.04	0.02	0.04
Labor	0.01	0.01	0.01	0.01
Other variable	0.04	0.05	0.05	0.04
Utilities	0.03	0.06	0.19	0.08
Materials	1.60	2.04	2.58	1.53
Profit ($/kg)	**2.7**	**5.57**	**10.87**	**97.89**