Supplementary Information

for

Synergetic pyrolysis of lithium-ion battery cathodes with polyethylene terephthalate for efficient metal recovery and battery regeneration

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Table S1 Loss of ignition (LOI) of the PET+ $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ **(**NCM) mixture during synergetic pyrolysis.

NCM:PET	Time (min)	Temperature (°C)	LOI* (%)
1:1	120	400	17.09
1:1	120	500	31.63
1:1	120	550	35.34
1:1	120	600	37.32
1:1	120	700	40.74
1:1	120	800	46.79
1:1	120	900	55.70
1:1	120	1000	61.06
1:0.3	15	600	12.32
1:0.3	30	600	17.74
1:0.3	60	600	17.88
1:0.3	90	600	17.68
1:0.3	120	600	17.90
1:0.1	120	600	8.00
1:0.2	120	600	11.99
1:0.3	120	600	17.89
1:0.4	120	600	24.04
1:0.5	120	600	26.31
1:1	120	600	37.32

*The mass of the feedstock before pyrolyzation and the products after pyrolyzation was recorded, and the loss of ignition (LOI) was calculated by dividing the mass difference between feedstock and products by the mass of feedstock.

** PET is polyethylene terephthalate.

Table S2 Comparison of the parameters of the thermal treatment of $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$

Technology	**Conditions**	**Products**	**Published year**	***Ref***
Carbothermal reduction roasting	Graphite, 650℃, 60min	Li_2CO_3, $(NiO)_m(MnO)_n$, Co, Ni	2020	1
Carbothermal reduction roasting	Graphite, 900℃, 90min	Li_2CO_3, MnO, Ni, Co	2021	2
Carbothermal reduction roasting	Graphite, 600℃, 180min	CoO, MnO, NiO, Li_2CO_3	2020	3
Sulfation roasting	$(NH_4)_2SO_4$, 650℃, 150min	Li_2CO_3, NiO, Co_3O_4, $LiMn_2O_4$	2020	4
Sulfation roasting	$NiSO_4$, 550℃, 60min	Li_2SO_4, Ni-Co-Mn-O	2022	5
Chlorination roasting	$CaCl_2$, 800℃, 60min	LiCl, NiO, CoO, MnO_2	2021	6
Chlorination roasting	Cl_2, 900℃, 90min	LiCl, $MnCl_2$, $CoCl_2$, $NiCl_2$	2022	7
Vacuum pyrolysis	Macadamia shell, 500℃+750℃,25min, Microwave roasting(2450MHZ)	Li_2CO_3, MnO, Ni-Co	2020	8

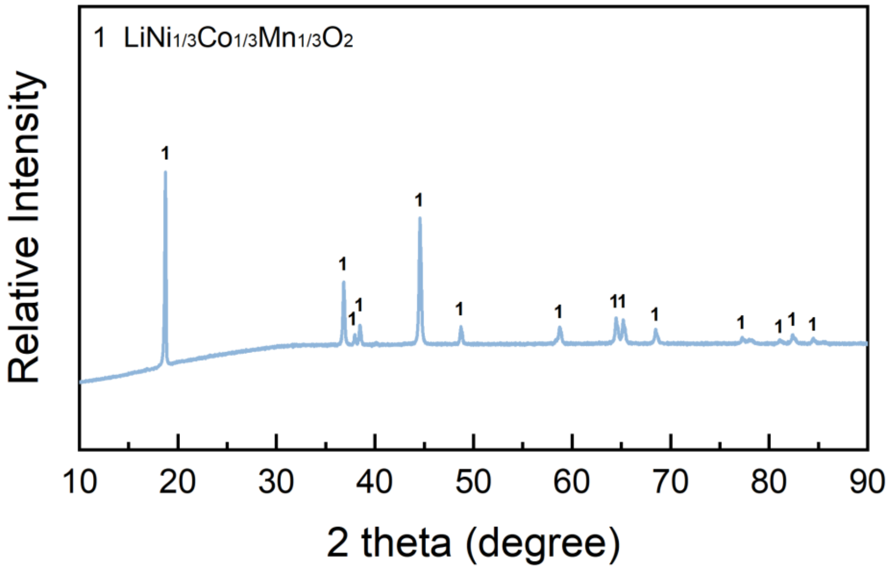

Figure S1 XRD patterns of the pyrolyzed $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ (NCM) without any additives under 600°C for 120 min.

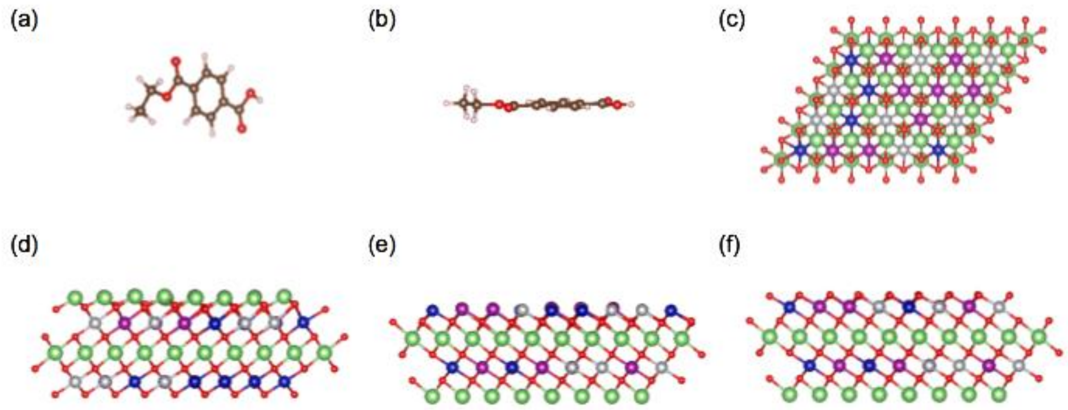

Figure S2 Structural diagrams of (a) NCM; (b) PET; (c) top view of NCM (001) slab; and front views of NCM (001) slab with (d) lithium, (e) transition metals, and (f) oxygen terminations. NCM is $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ and PET is polyethylene terephthalate.

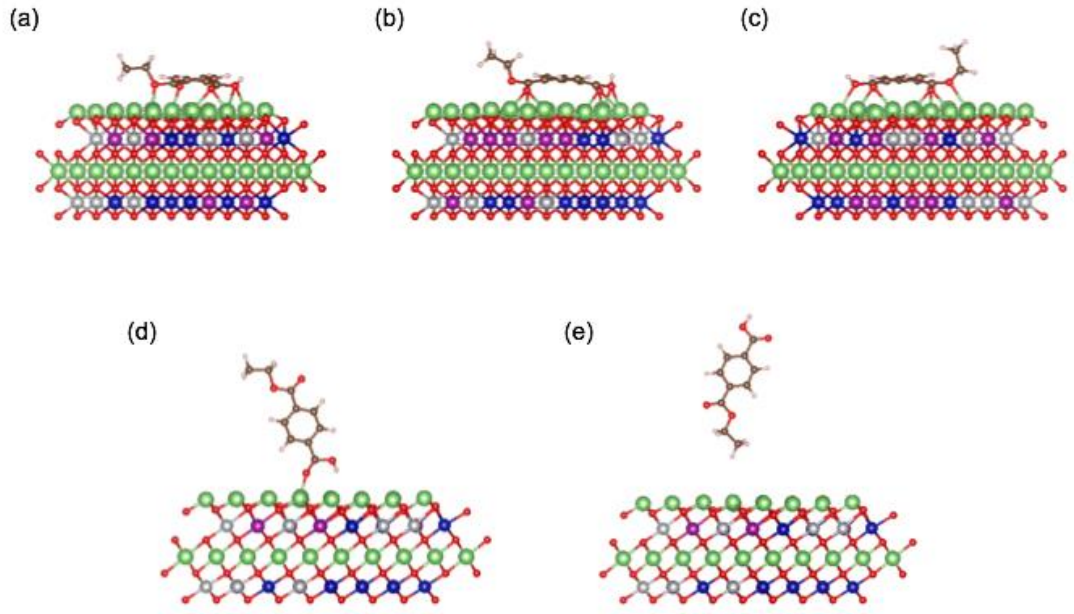

Figure S3 Adsorption configurations of PET on NCM (001) slab with lithium terminations. Front views of (a) Configuration 1, (b) Configuration 2, (c) Configuration 3, (d) Configuration 4 and (e) Configuration 5. NCM is $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ and PET is polyethylene terephthalate.

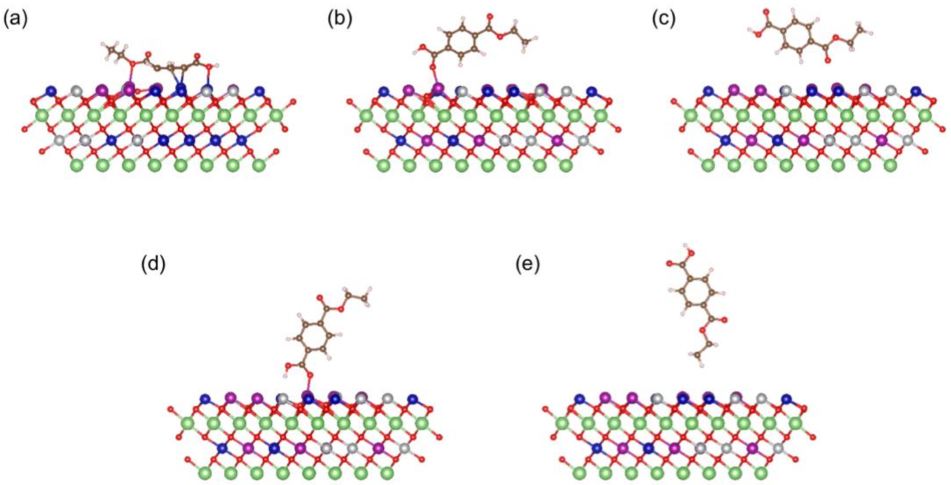

Figure S4 Adsorption configurations of PET on NCM (001) slab with transition metal terminations. Front views of (a) Configuration 6, (b) Configuration 7, (c) Configuration 8, (d) Configuration 9 and (e) Configuration 10. NCM is $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ and PET is polyethylene terephthalate.

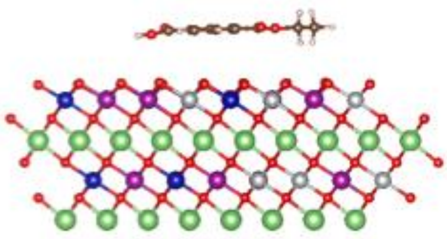

Figure S5 Adsorption configuration of PET on NCM (001) slab with oxygen terminations. (a) front view of Configuration 11. NCM is $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ and PET is polyethylene terephthalate.

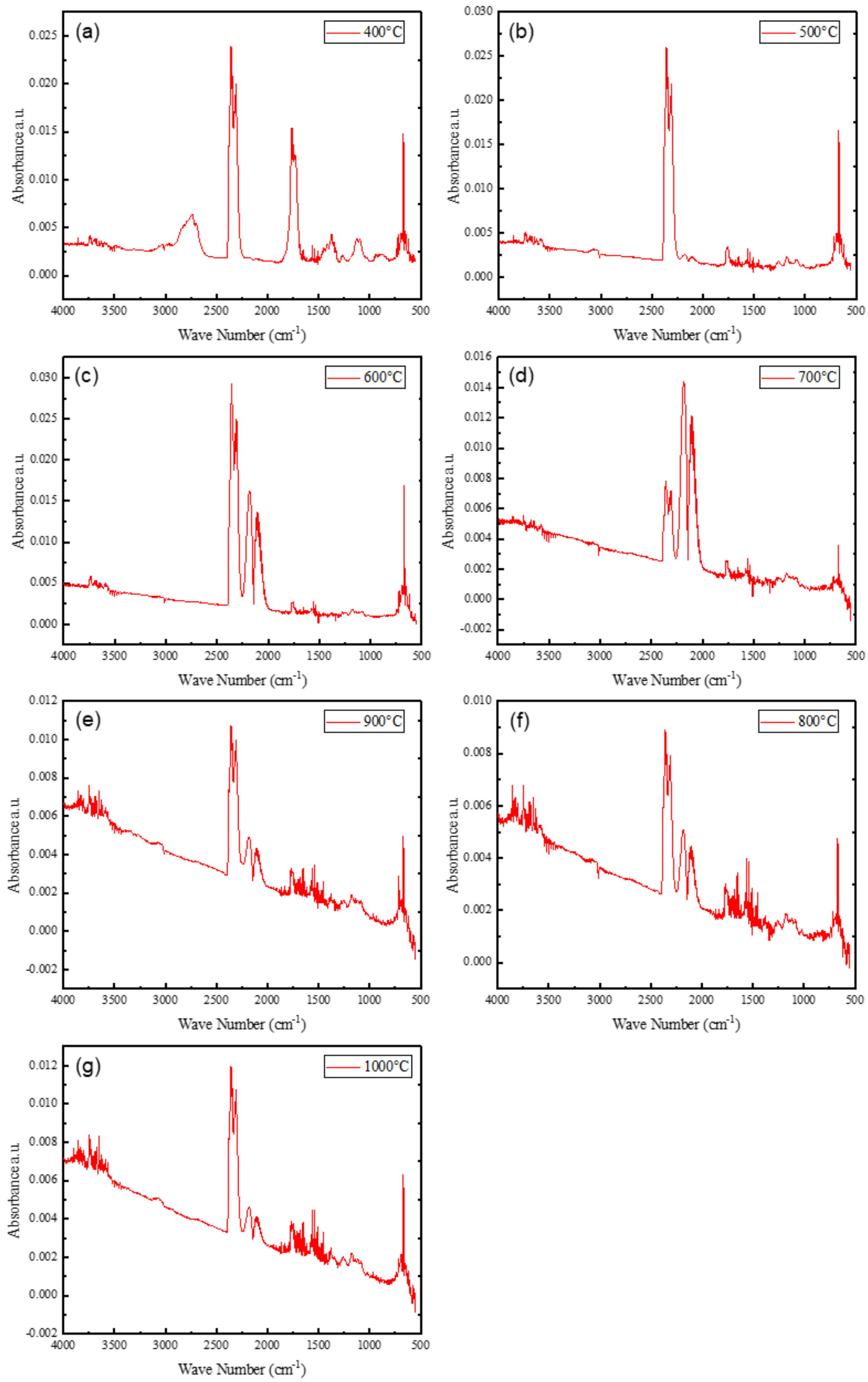

Figure S6 FT-IR spectra at different pyrolysis temperatures. (a) 400°C, (b) 500°C, (c) 600°C, (d) 700°C, (e) 800°C, (f) 900°C, (g) 1000°C.

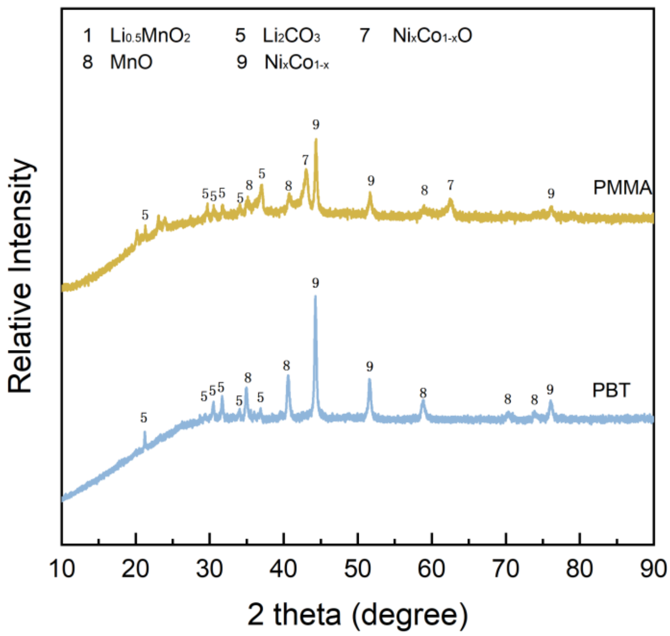

Figure S7 Typical XRD patterns of the plastic + NCM ($LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$) synergetic pyrolysis products at 550°C for 60 min with a mass ratio of NCM/plastic of 1.0:0.5.

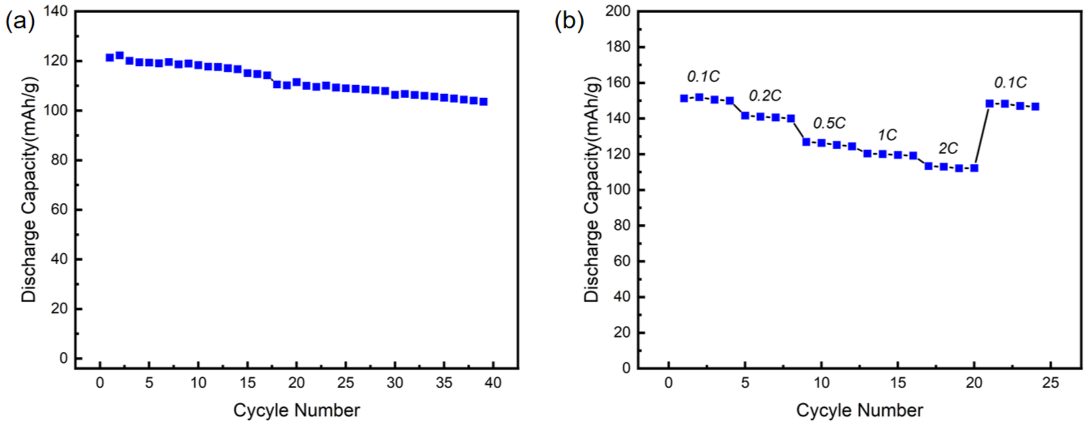

Figure S8 (a) Cycle performance at 1C and (b) rate performance of Sample R-NCM, where R-NCM is regenerated $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$.

Supplementary References

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