Electronic Supplementary Material (ESI) for Dalton Transactions.

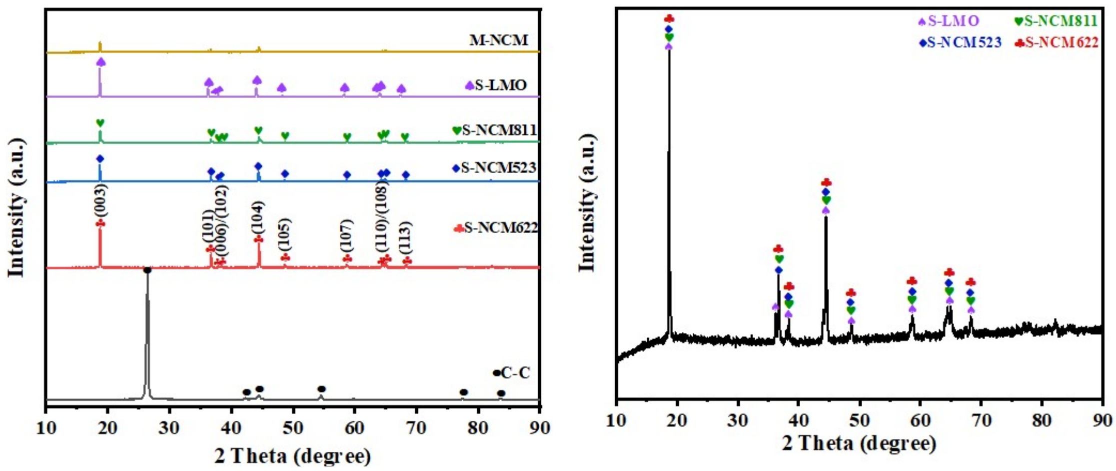




Figure S1 XRD of Commercial graphite（C-C）、$LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$（S-NCM622）、$LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$（S-NCM523）、$LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$（S-NCM811）、$LiMn_2O_4$（S-LMO）and mixture NCM（M-NCM）

Table S1 ICP test results of unburned mixed raw materials

| Elements | Li | Ni | Co | Mn |
|---|---|---|---|---|
| Wt.（%） | 6.74 | 7.34 | 27.83 | 22.65 |

S 2.2 Handling the emitted $CO_2$ in the experimental process:

Inject lime water into a conical flask with a capacity of 2/3 and seal with a cork. Use a rubber tube to pass through the cork of the conical bottle, connecting one end to the outlet end of the tube furnace and the other end to the conical bottle. Place the rubber tube placed at this end of the conical flask 1/3 below the liquid level of the lime water solution.

At the beginning of the reaction, the roasting process will produce a small amount of $CO_2$ entering the lime water，the reaction is as follows (2-1):

$$Ca(OH)_2+CO_2 = CaCO_3 + H_2O \qquad (2\text{-}1)$$

Produce $CaCO_3$ precipitation, and the lime water becomes turbid. With the temperature increases (700 ℃-800 ℃), excessive $CO_2$ is introduced into the turbid lime water, which will continue to react (2-2):

$$CaCO_3 + CO_2 + H_2O = Ca(HCO_3)_2 \qquad (2\text{-}2)$$

The precipitation dissolves and the solution become clear.‘’

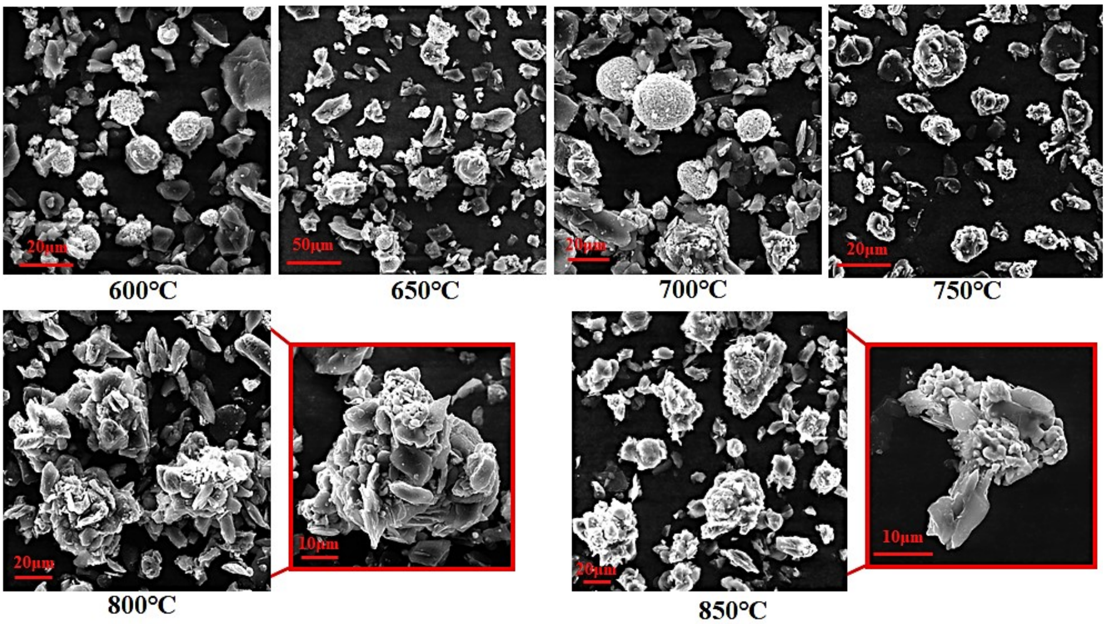


Figure S2. SEM of calcined raw materials at different calcination temperatures (roasting for 3 h, mixture cathode materials: graphite=1:1)

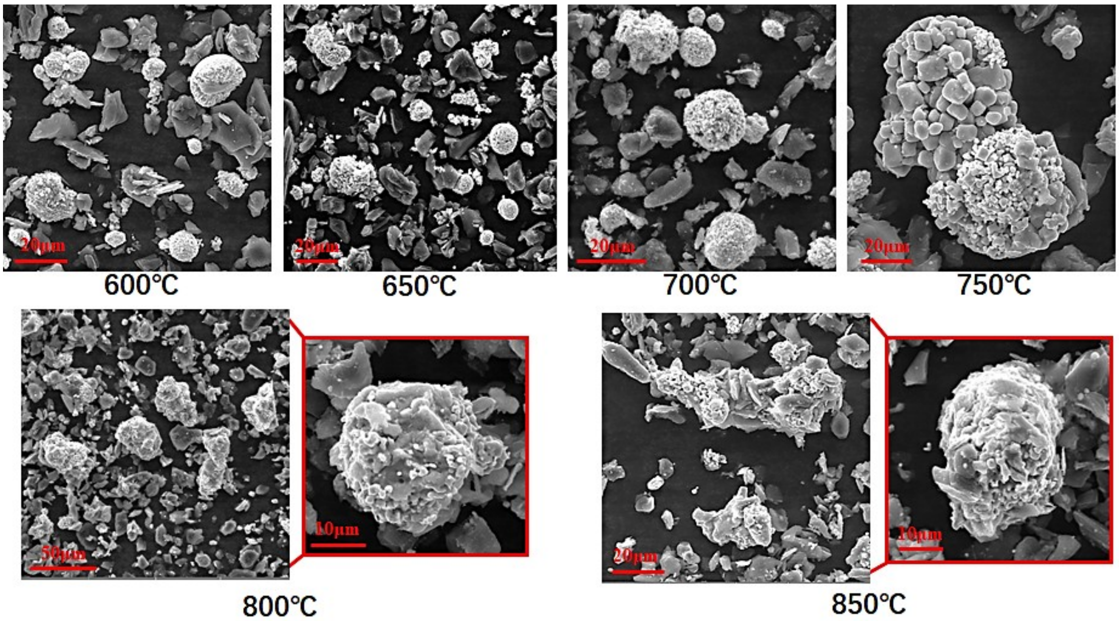
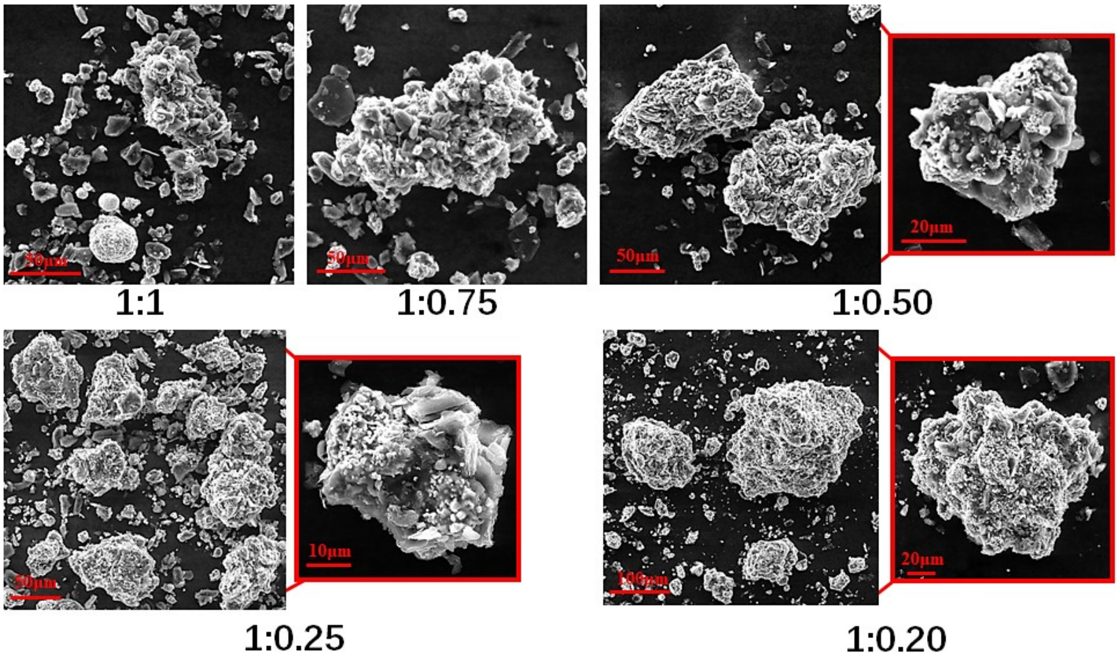
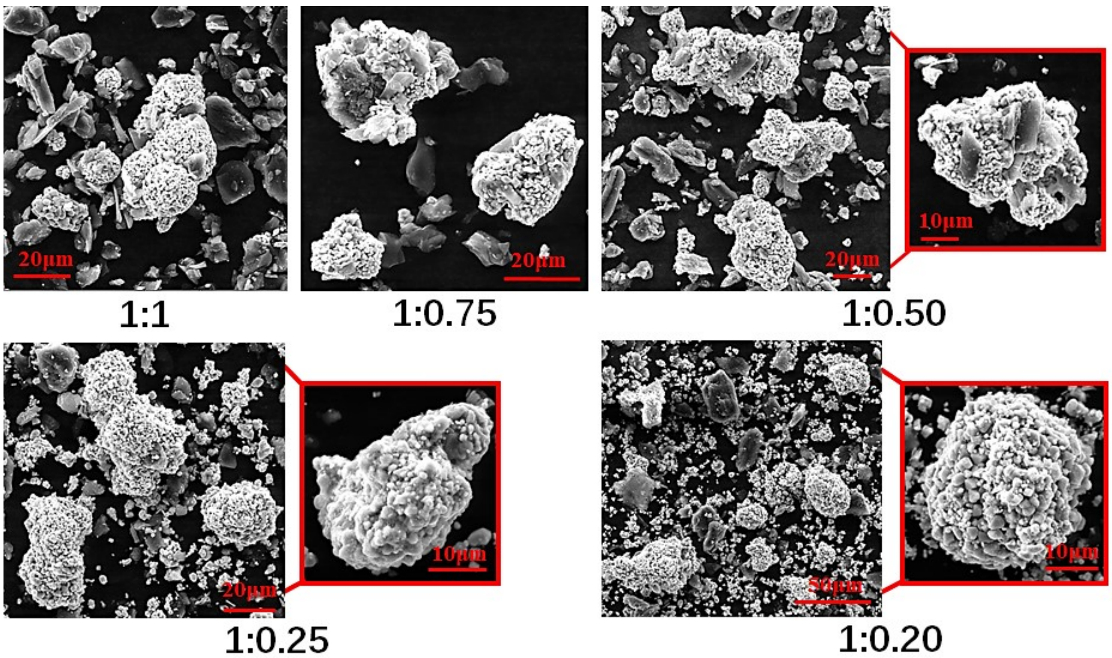


Figure S3. SEM of filter residue materials at different calcination temperatures (roasting for 3 h, mixture cathode materials: commercial graphite=1:1)




FigureS4. SEM of Raw Materials Diagram of Roasting Products with Different Material Proportions (roasting temperature750°C,3h)




FigureS5. SEM of Filter residue Materials Diagram of Roasting Products with Different Material Proportions (roasting temperature750°C,3h)

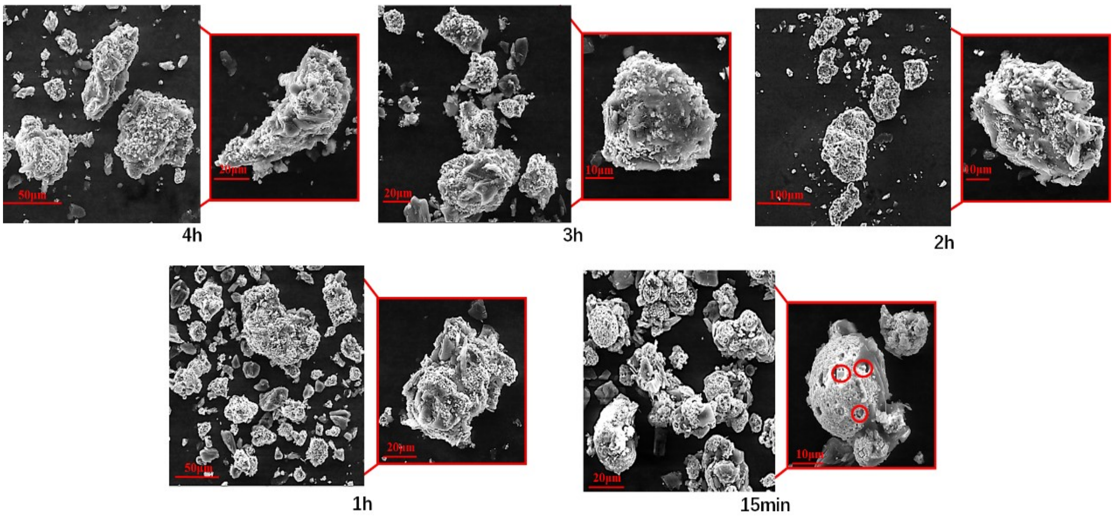



Figure S6. SEM of Raw materials diagram of calcination products at different times (roasting temperature 750h, mixed cathode material: graphite=1:0.25)

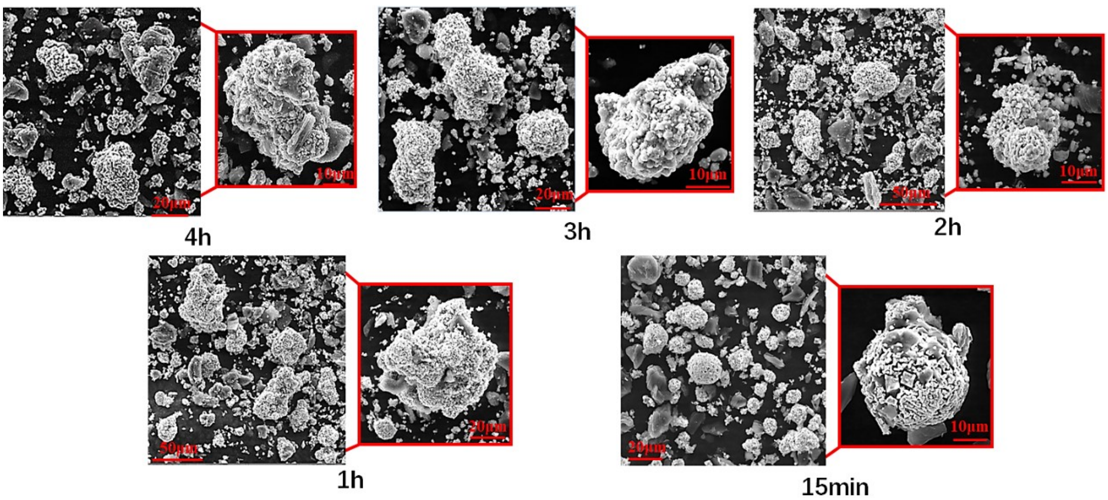



Figure S7. SEM of Filter residue materials diagram of calcination products at different times (roasting temperature 750h, mixed cathode material: graphite=1:0.25)

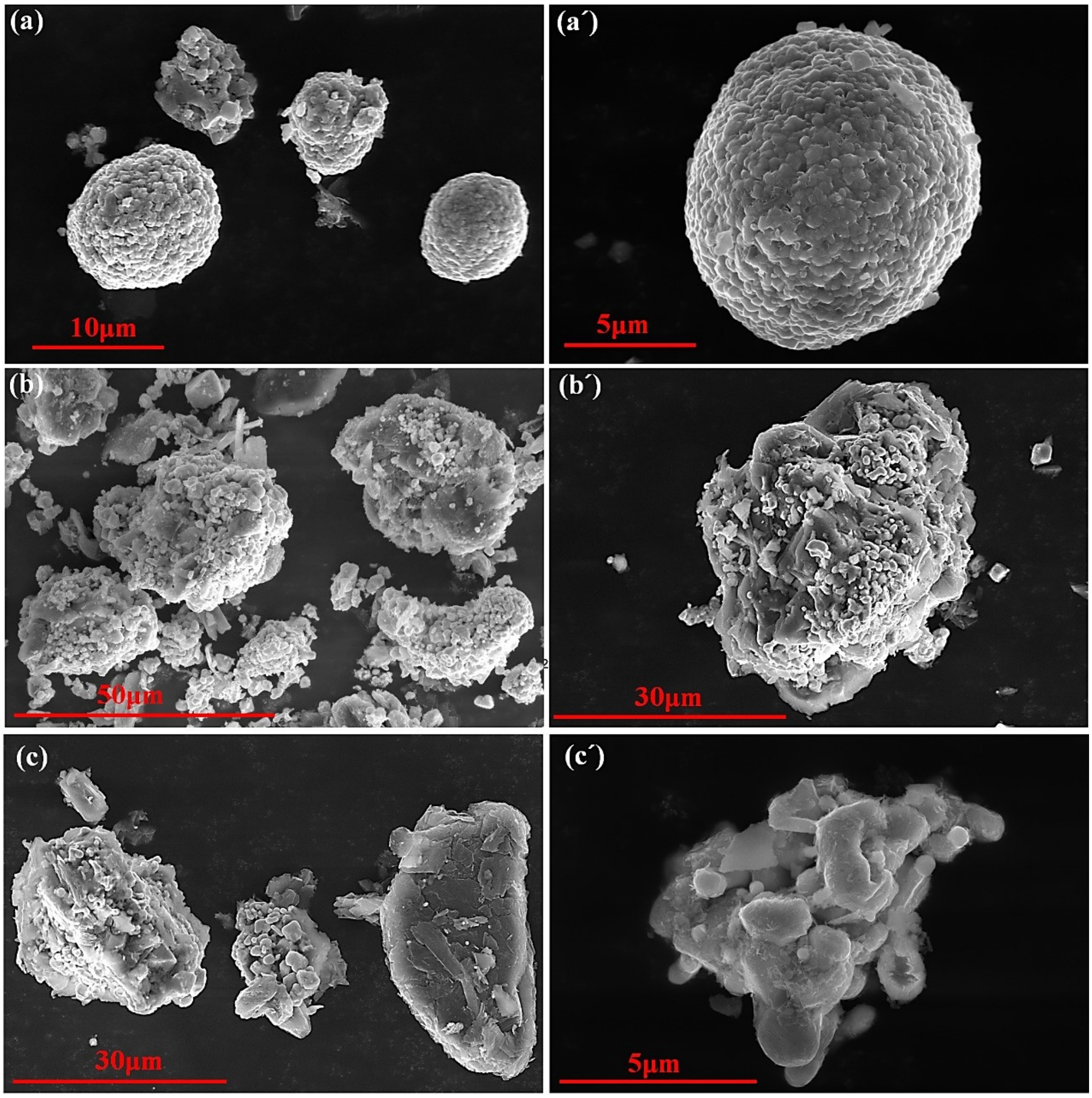



Figure S8. Roasting temperature 750 ℃, insulation for 4 h, mixture cathode materials: graphite=1:0.25（a-a´）Unburned raw materials; (b-b´) Raw material（c-c´）Filter residue materials

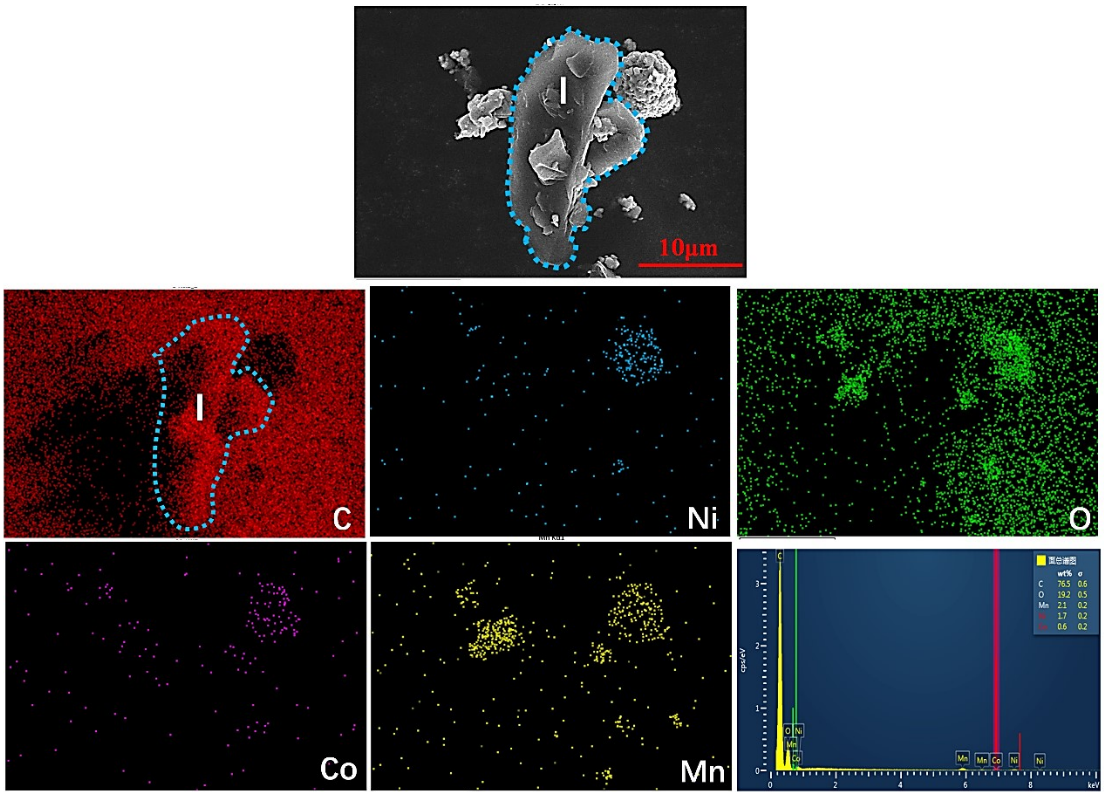


Figure S9. TEM of mixture cathode materials: carbon=1:0.25 without roast

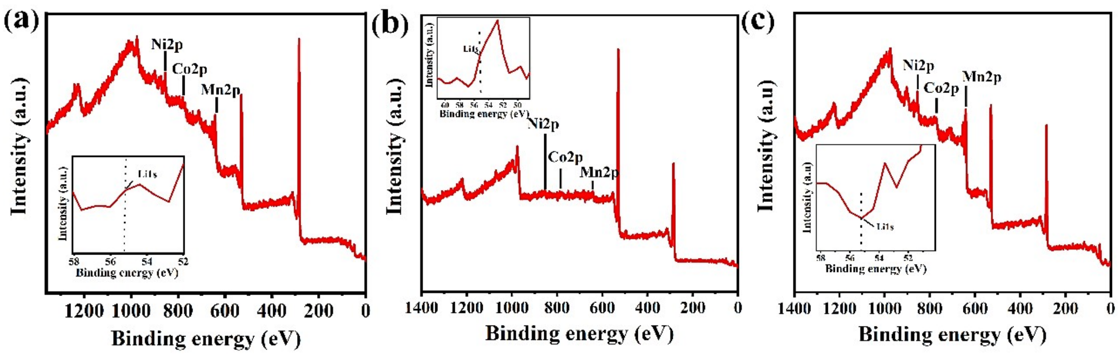



Figure S10. XPS of（a）Unburned raw materials; (b) Raw material（c）Filter residue materials

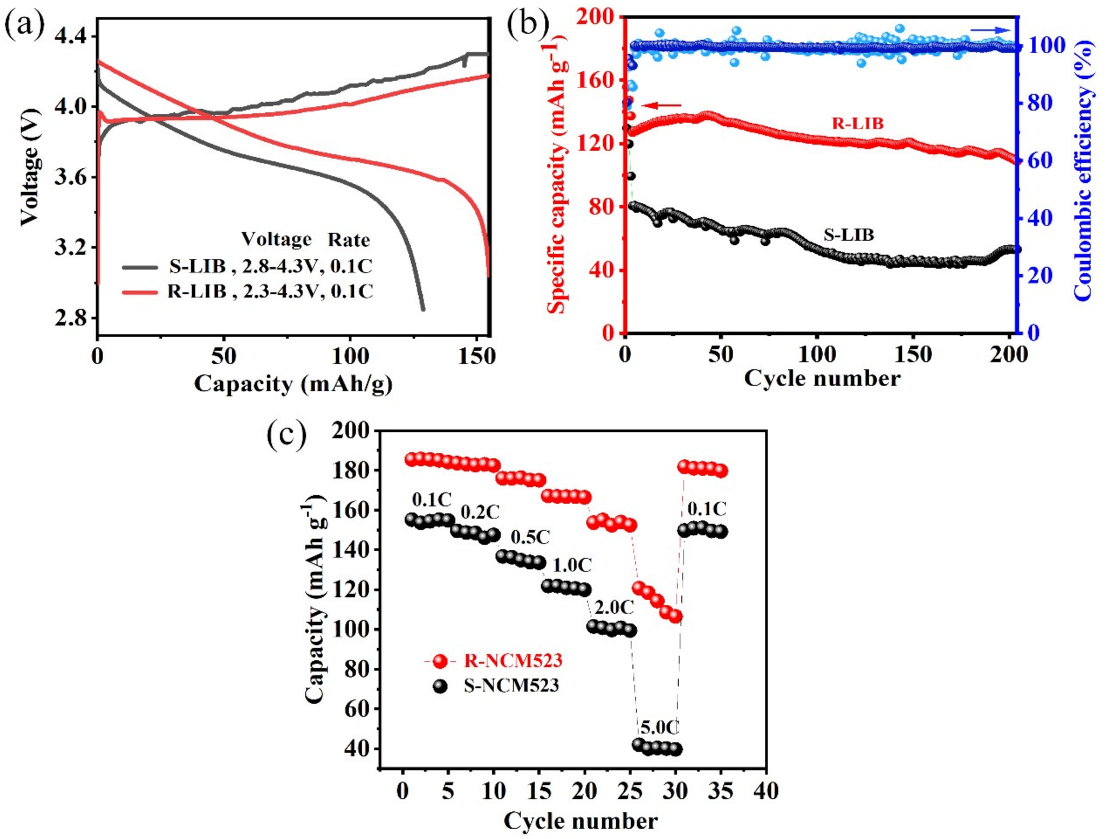



Figure S11. The first cycle of charge and discharge at (a) 0.1C and (b) 200 cycles 1C at 2.8-4.3V for the waste positive electrode material (S-LIB) and the cathode material (R-LIB) directly regenerated from the extracted $Li_2CO_3$ as a lithium source (R-LIB); the rate performance diagram of(c)spent $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$(S-NCM523) and using $Li_2CO_3$ extracted in this article as a lithium source to repair waste NCM523 batteries(R-NCM523).