

## Supporting Information

### **A closed-loop regeneration of $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ and graphite from spent battery via efficient lithium supplement and structural remodelling**

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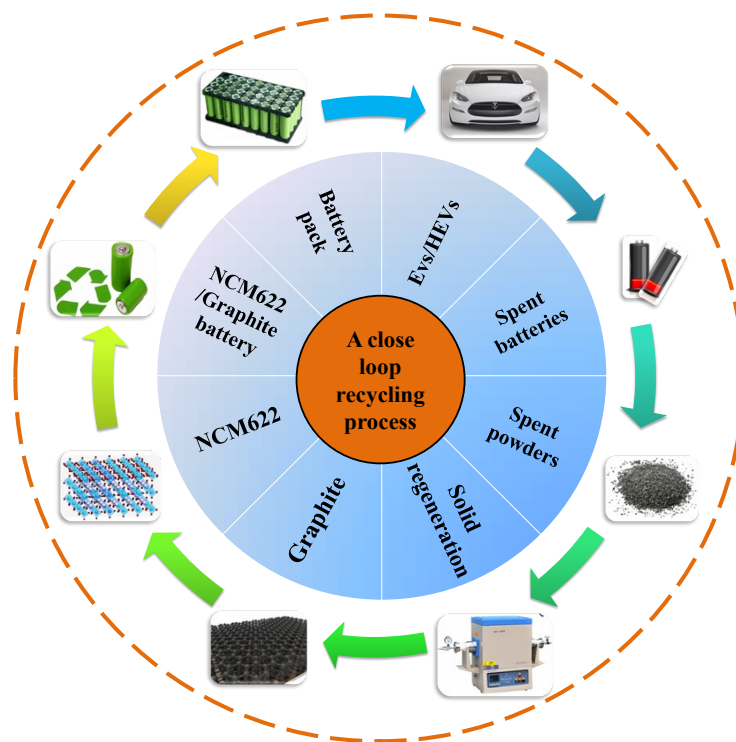
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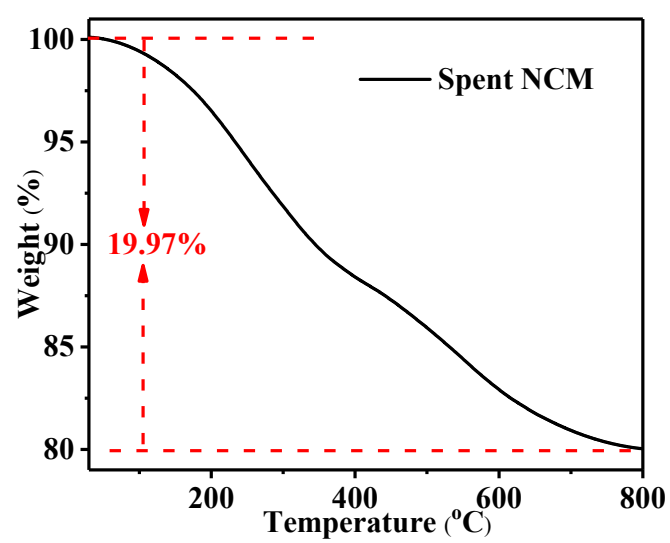
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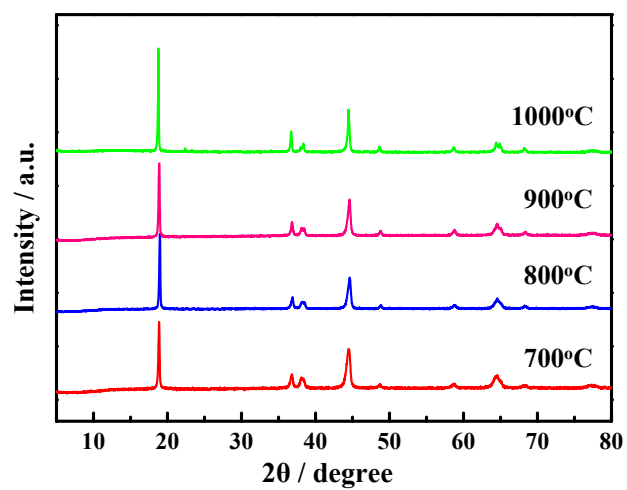
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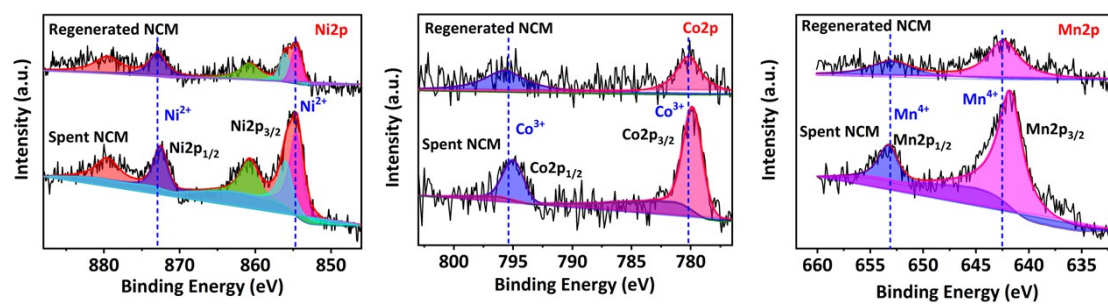
**Fig. S1.** The whole recycling and regeneration processes of NCM622 and graphite in this research.



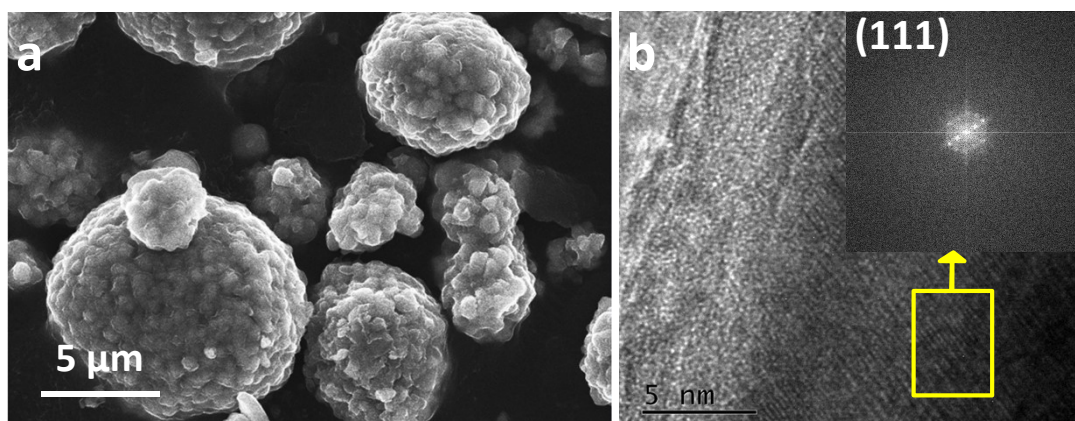
**Fig. S2.** TG curves of spent NCM622 sample in this experiment.



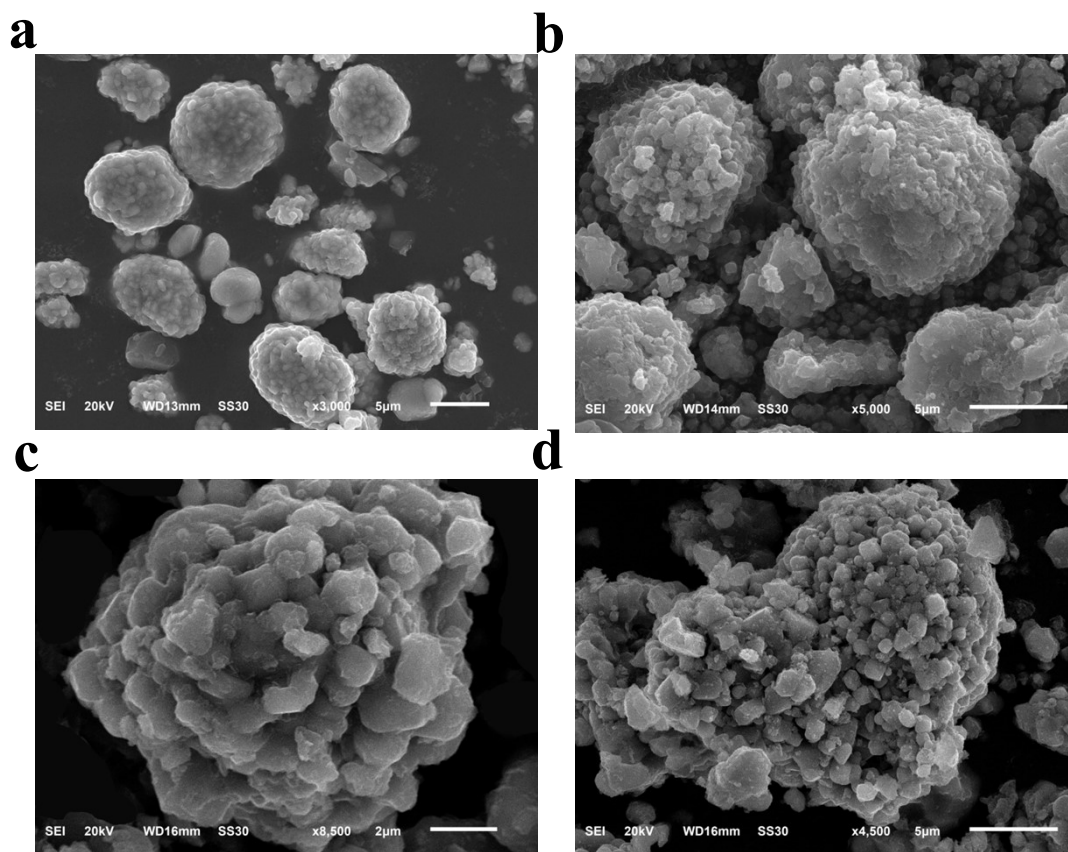
**Fig. S3.** XRD patterns of regenerated NCM samples at different temperatures.



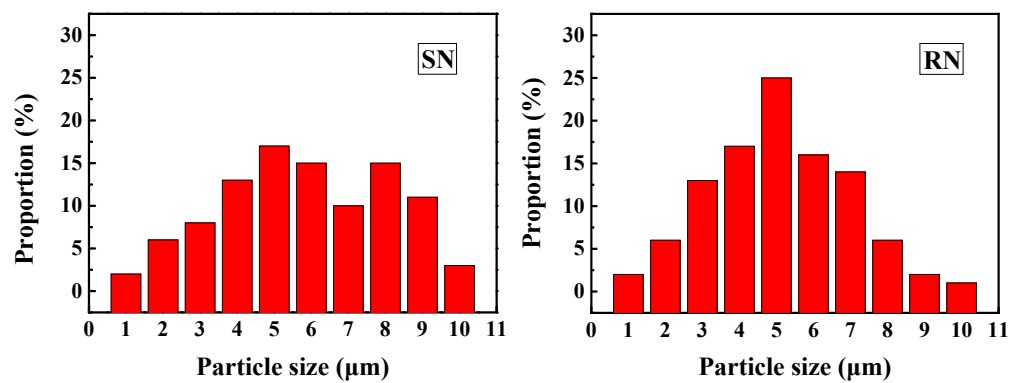
**Fig. S4.** XPS spectra of Ni2p, Co2p and Mn2p in the spent and regenerated NCM622 samples.



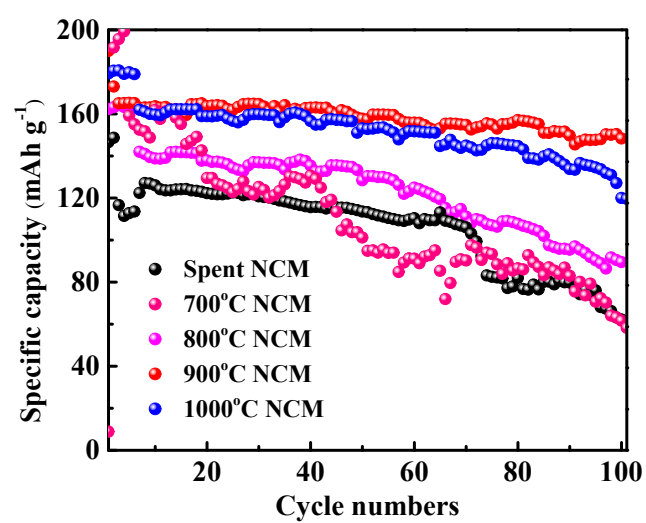
**Fig. S5.** (a) SEM and (b) HRTEM images of spent NCM622 sample.



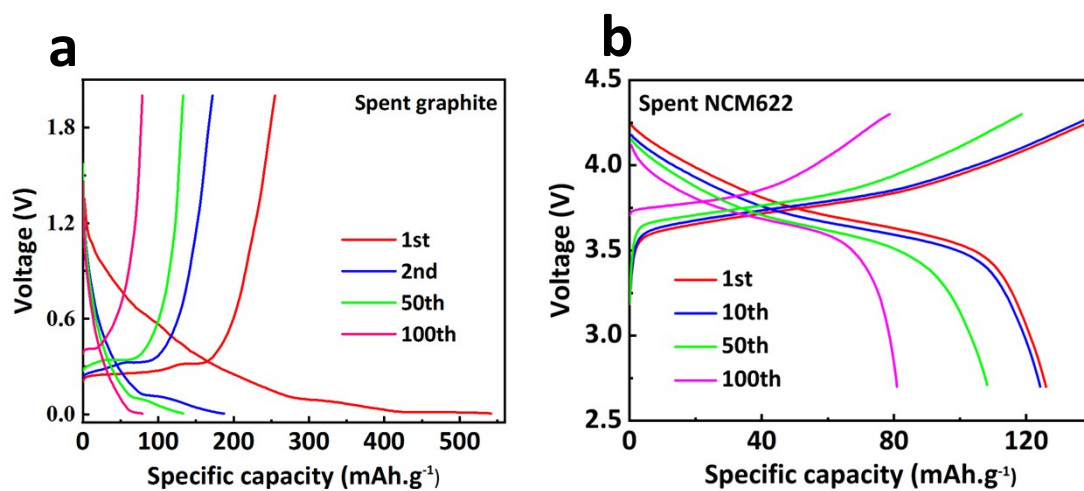
**Fig. S6.** SEM images of RN regenerated at (a) 700, (b) 800, (c) 900 and (d) 1000 °C.



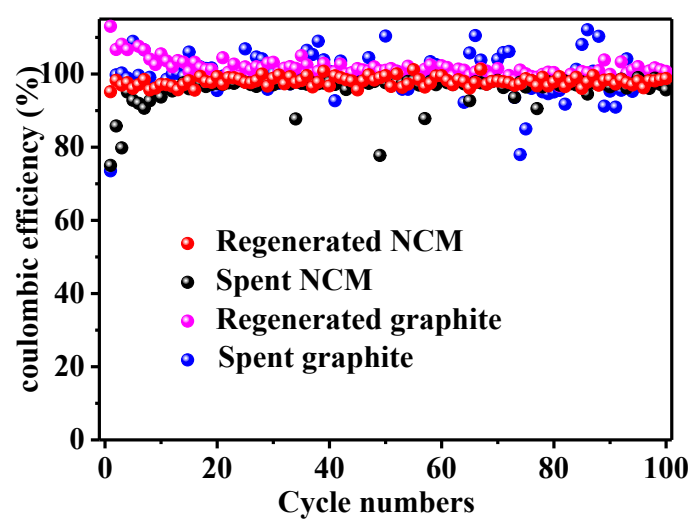
**Fig. S7.** Particle distributions of spent NCM622 and regenerated NCM622 samples (regenerated at optimal temperature).



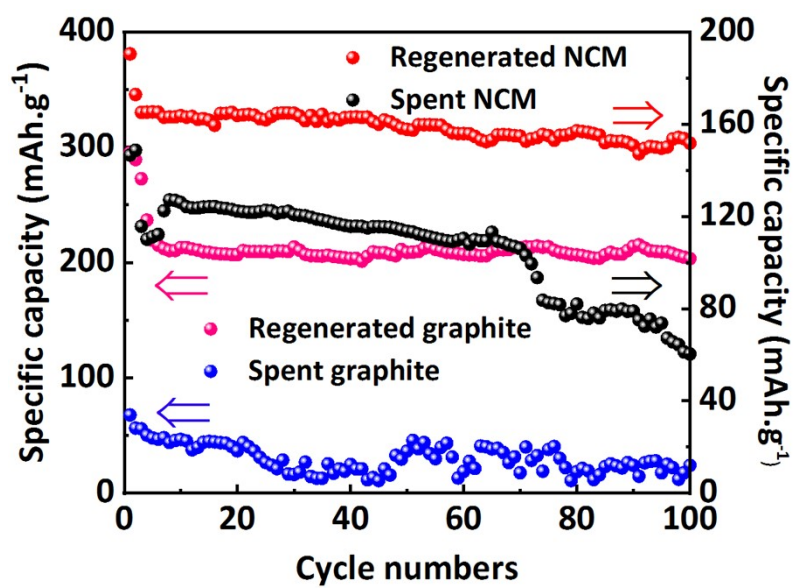
**Fig. S8.** Cycling performances of NCM materials regenerated at different temperatures.



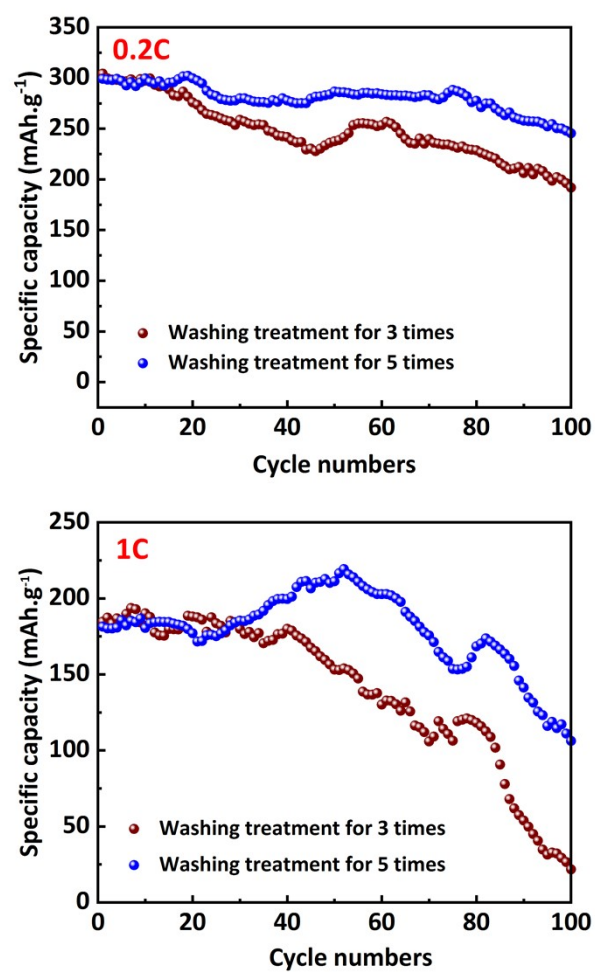
**Fig. S9.** Charge-discharge profiles of (a) spent graphite and (b) spent NCM622 half batteries at 0.2C in 1st, 10th, 50th and 100th cycle.



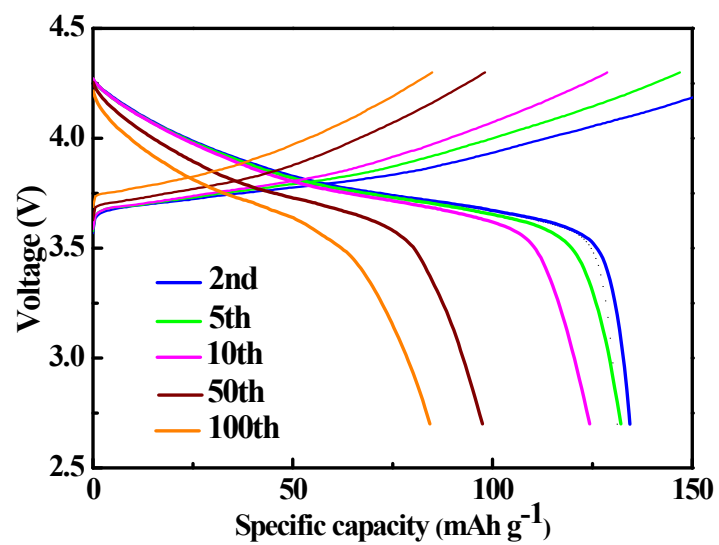
**Fig. S10.** Coulombic efficiency of RN, SN, RG and SG electrodes at 0.2C.



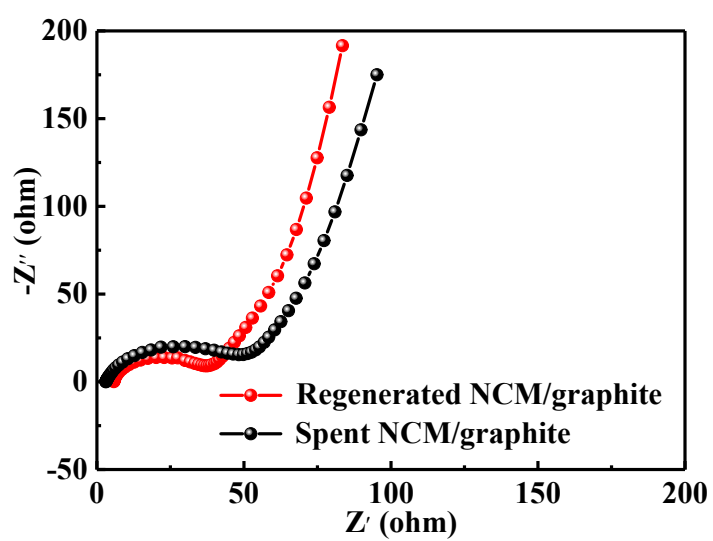
**Fig. S11.** Cycling performances of regenerated NCM622 and regenerated graphite half batteries at 1C.



**Fig. S12.** Cycling performance of spent graphite electrodes after washing for 3 or 5 times.



**Fig. S13.** Charge-discharge profiles of SN/SG full battery in the potential range of 2.7-4.3 V at 0.2C in 2nd, 5th, 10th, 50th and 100th cycle.



**Fig. S14.** Electrochemical impedance spectra of RN/RG and SN/SG full cells with an amplitude of 5.0 mV over the frequency range of 100 kHz-0.01Hz after the first cycle.

**Table S1.** Metal contents including Li, Ni, Co and Mn calculated based on ICP results of spent and regenerated NCM622.

| Sample      | Li    | Ni    | Co    | Mn    |
|-------------|-------|-------|-------|-------|
| Spent NCM   | 0.635 | 0.614 | 0.215 | 0.192 |
| RN (T-700)  | 0.971 | 0.659 | 0.208 | 0.201 |
| RN (T-800)  | 0.995 | 0.610 | 0.210 | 0.203 |
| RN (T-900)  | 1.000 | 0.611 | 0.212 | 0.205 |
| RN (T-1000) | 1.000 | 0.610 | 0.211 | 0.204 |

**Table S2.**  $I_{(006)}/I_{(012)}$  and  $I_{(018)}/I_{(110)}$  values of XRD patterns from spent and regenerated NCM622 at different temperatures.

| Sample                | T-700 | T-800 | T-900 | T-1000 | Un-treated |
|-----------------------|-------|-------|-------|--------|------------|
| $I_{(006)}/I_{(012)}$ | 1.89  | 1.88  | 2.12  | 2.03   | 1.65       |
| $I_{(018)}/I_{(110)}$ | 1.06  | 1.07  | 1.13  | 1.10   | 0.89       |