

Supplementary material

Green Energy Storage: Upcycling of Lithium-ion Battery for its Utilization in a High-Performance Asymmetric Supercapacitor Device

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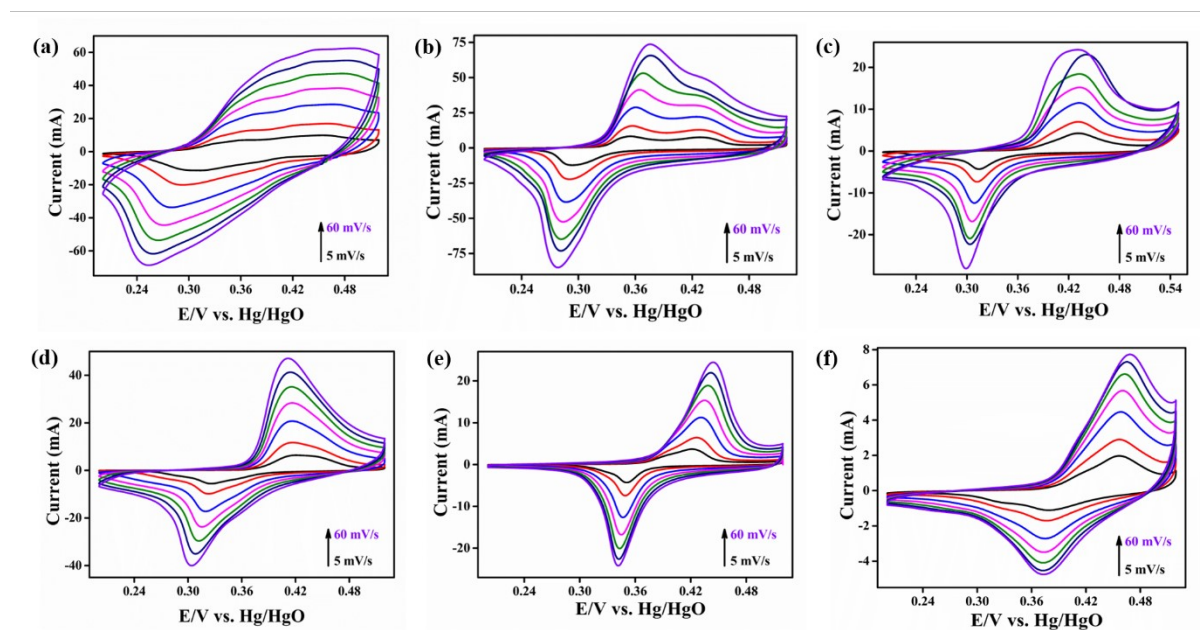


Figure S1 CV at increasing scan rate of (a) LCO-BS_NF; (b) LCO-S_NF; (c) LCO-C_NF; (d) GR-C_NF; (e) GR-S_NF; (f) GR-BS_NF

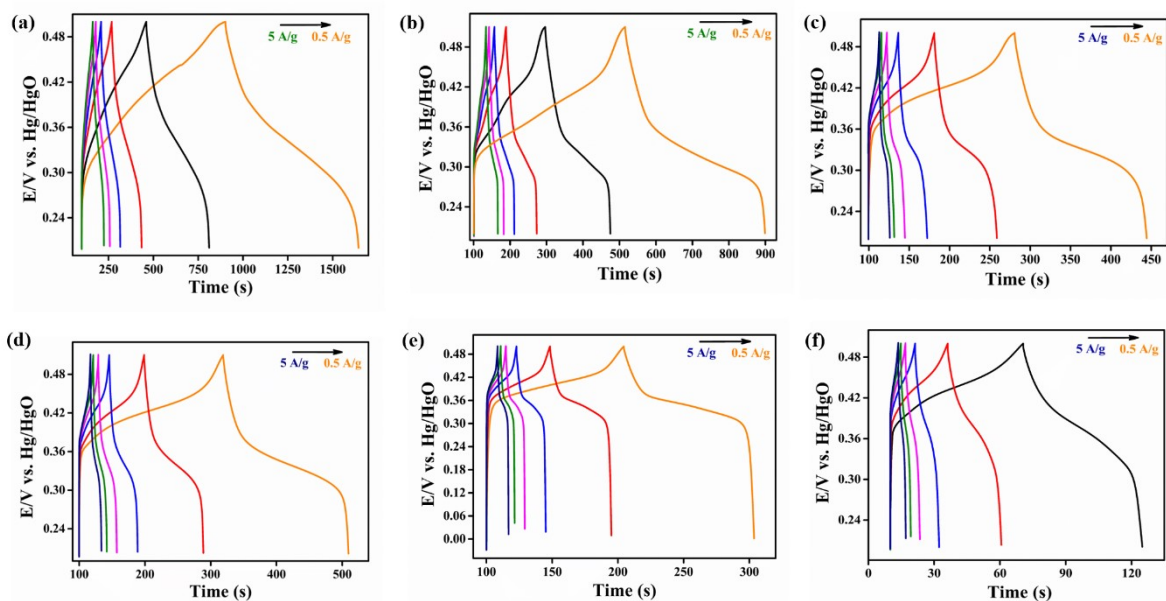


Figure S2 GCD at increasing current density of (a) LCO-BS_NF; (b) LCO-S_NF; (c) LCO-C_NF; (d) GR-C_NF; (e) GR-S_NF; (f) GR-BS_NF

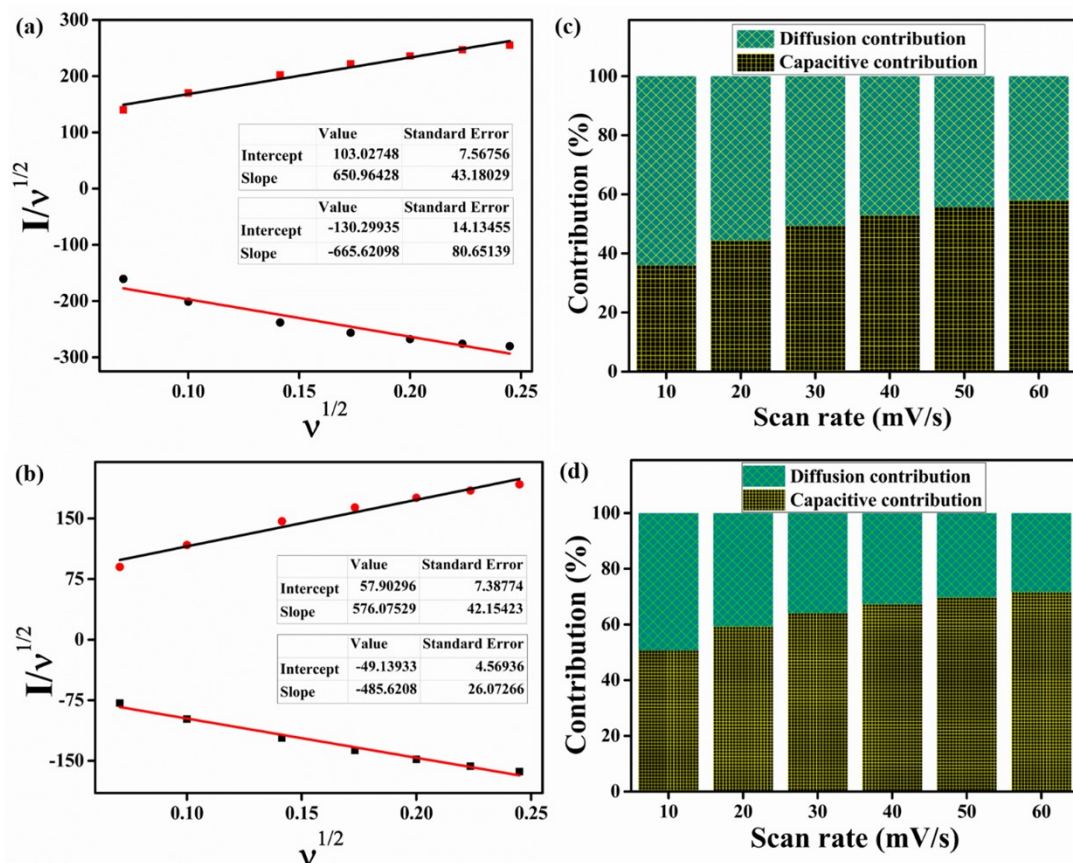


Figure S3 $I/v^{1/2}$ versus $v^{1/2}$ plot for oxidation and reduction current peak of (a) LCO-BS_NF and (b) GR-C_NF, and relative capacitive and diffusion contribution graphs of (c) LCO-BS_NF and (d) GR-C_NF.

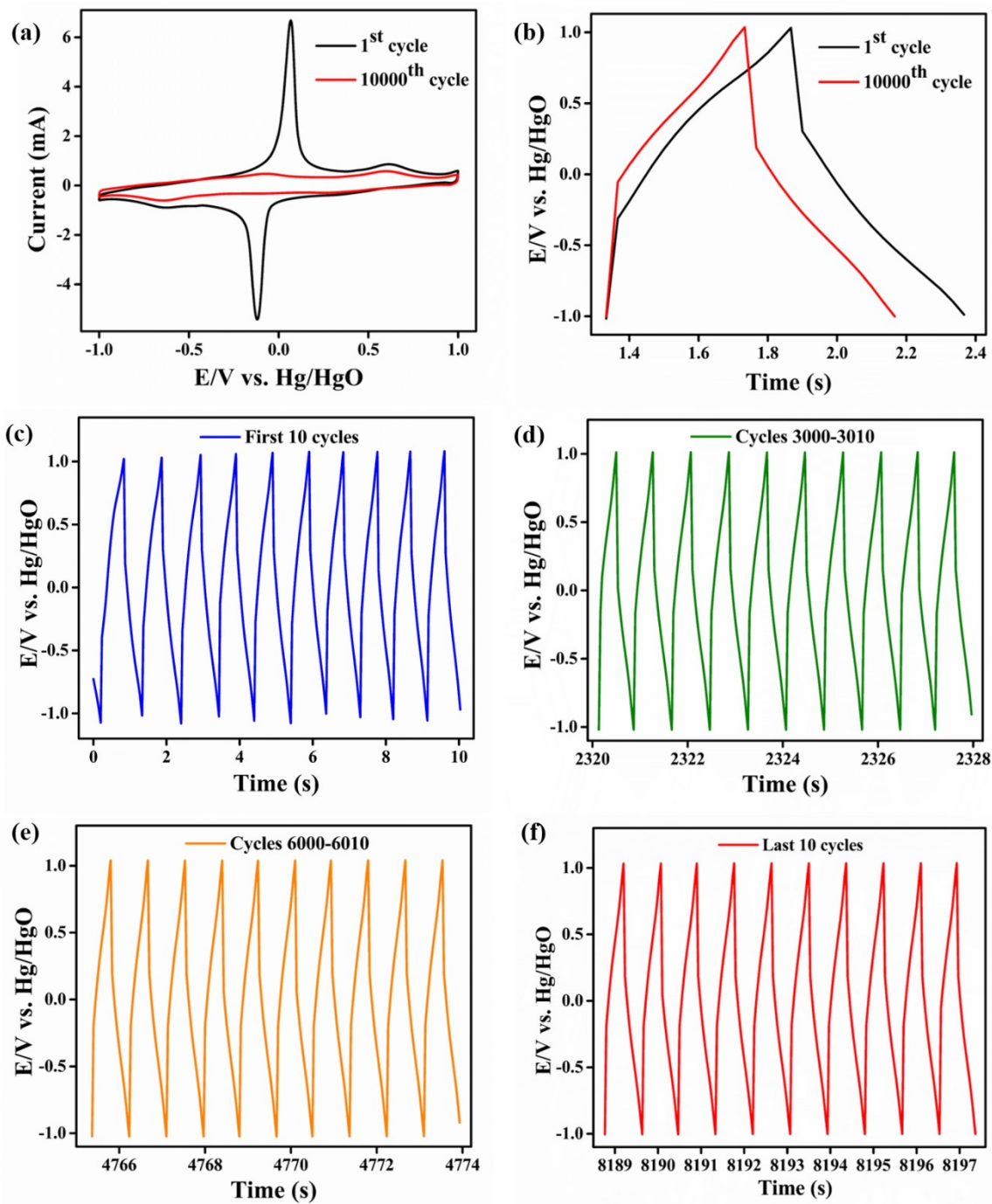


Figure S4 (a) Comparison of CV curve before and after cyclic stability; (b) GCD curve of the first cycle and the last cycle after cyclic stability; (c) first ten cycles of cyclic stability; (d),(e) intermediate cycles of cyclic stability, and (f) last ten cycles of cyclic stability

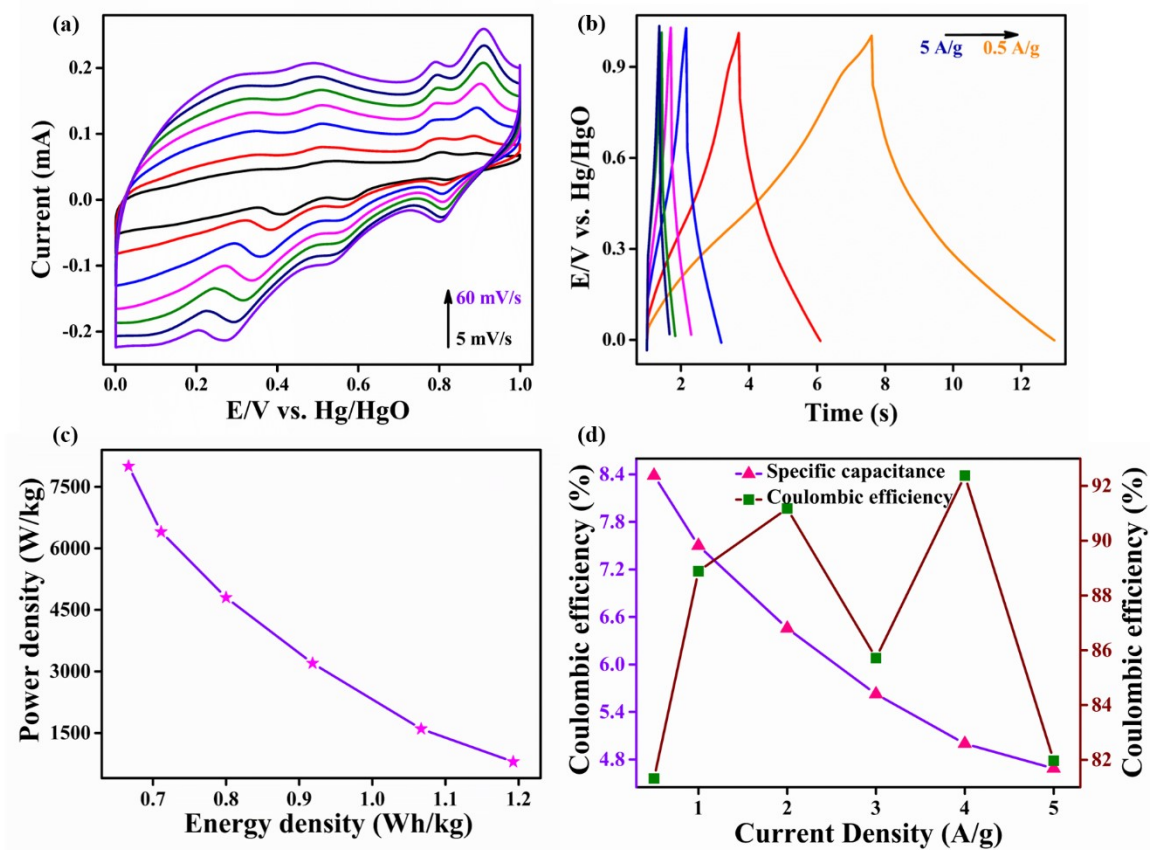


Figure S5 Symmetric device of LCO-BS_NF (a) CV at increasing scan rate; (b) GCD at increasing current density; (c) power density vs energy density curve and (d) graph showing specific capacitance and coulombic efficiency at various current densities

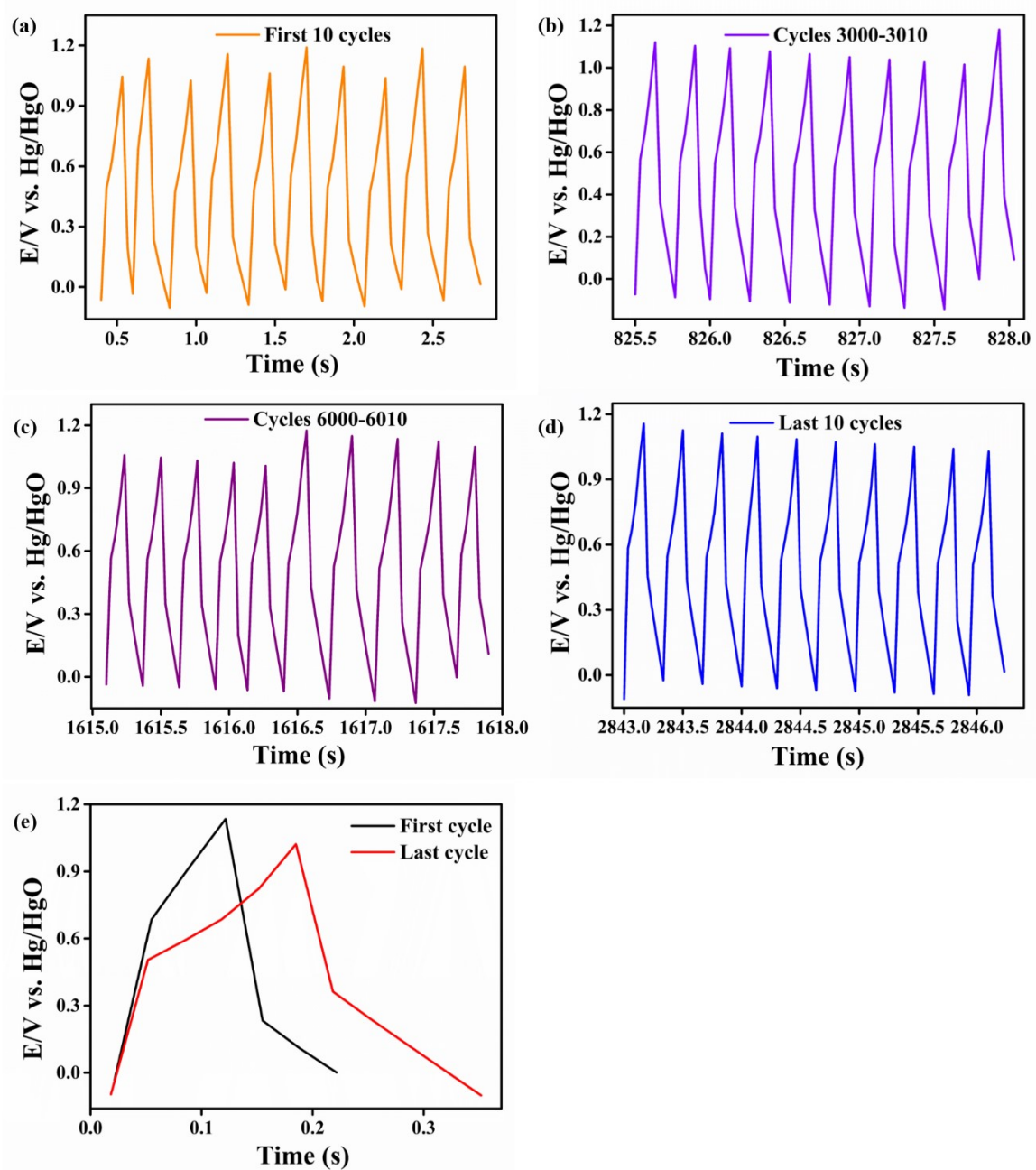


Figure S6 Symmetric device of LCO-BS_NF (a) first ten cycles of cyclic stability; (b),(c) intermediate cycles of cyclic stability; (d) last ten cycles of cyclic stability and (e) GCD curve of the first cycle and the last cycle after cyclic stability

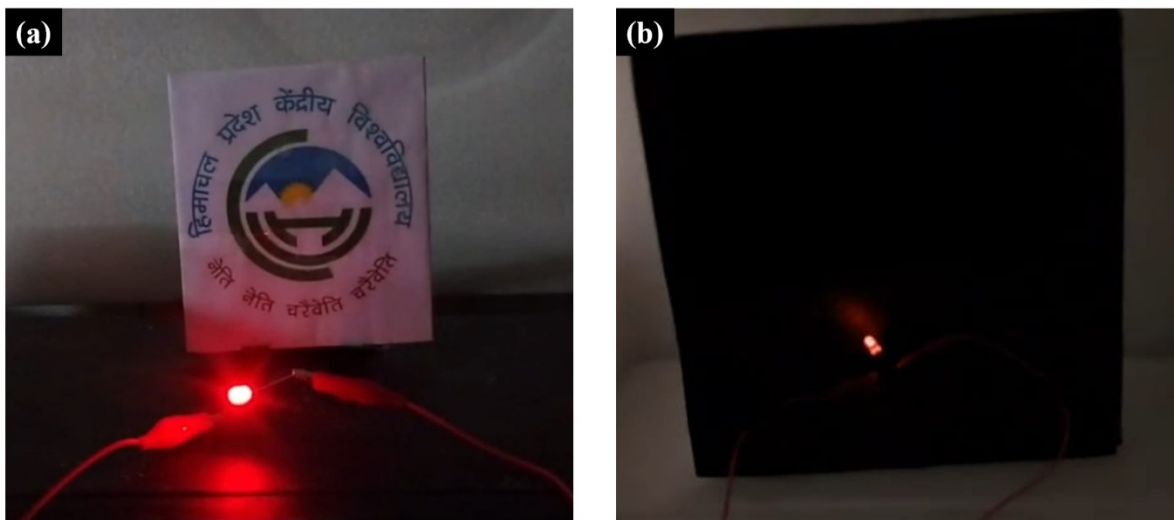


Figure S7 Illumination of red LED bulb by (a) symmetric LCO-BS_NF device and (b) asymmetric LCO-BS_NF and GR-C_NF device

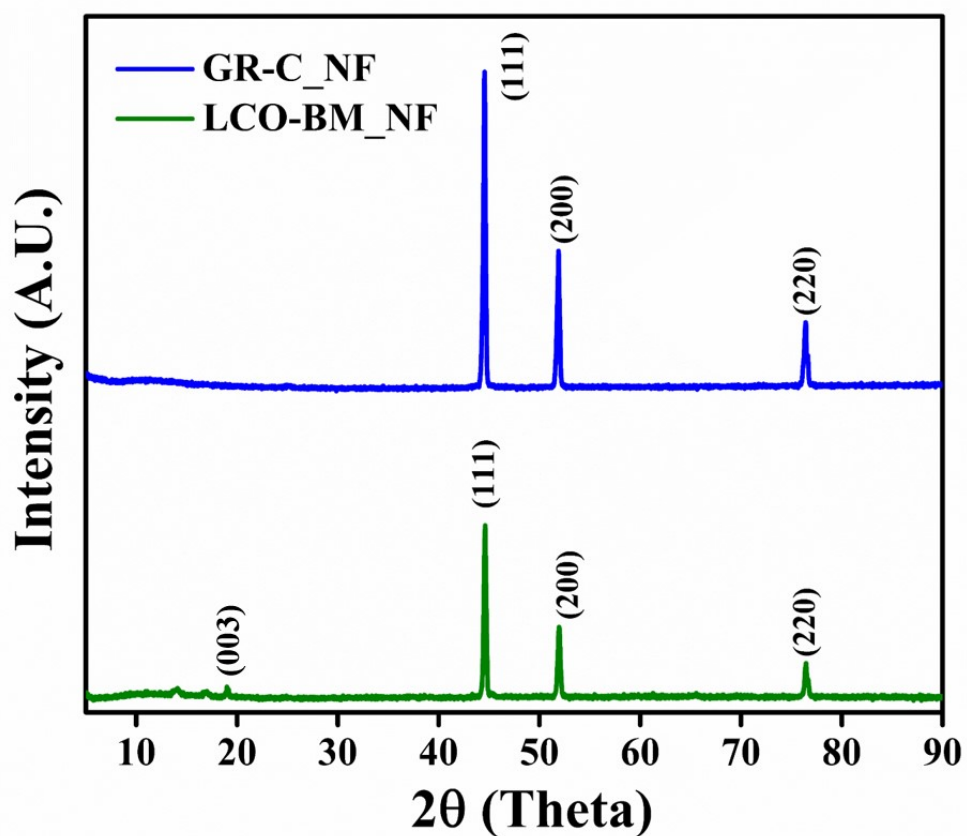


Figure S8 XRD diffractograms of GR-C_NF and LCO-BM_NF

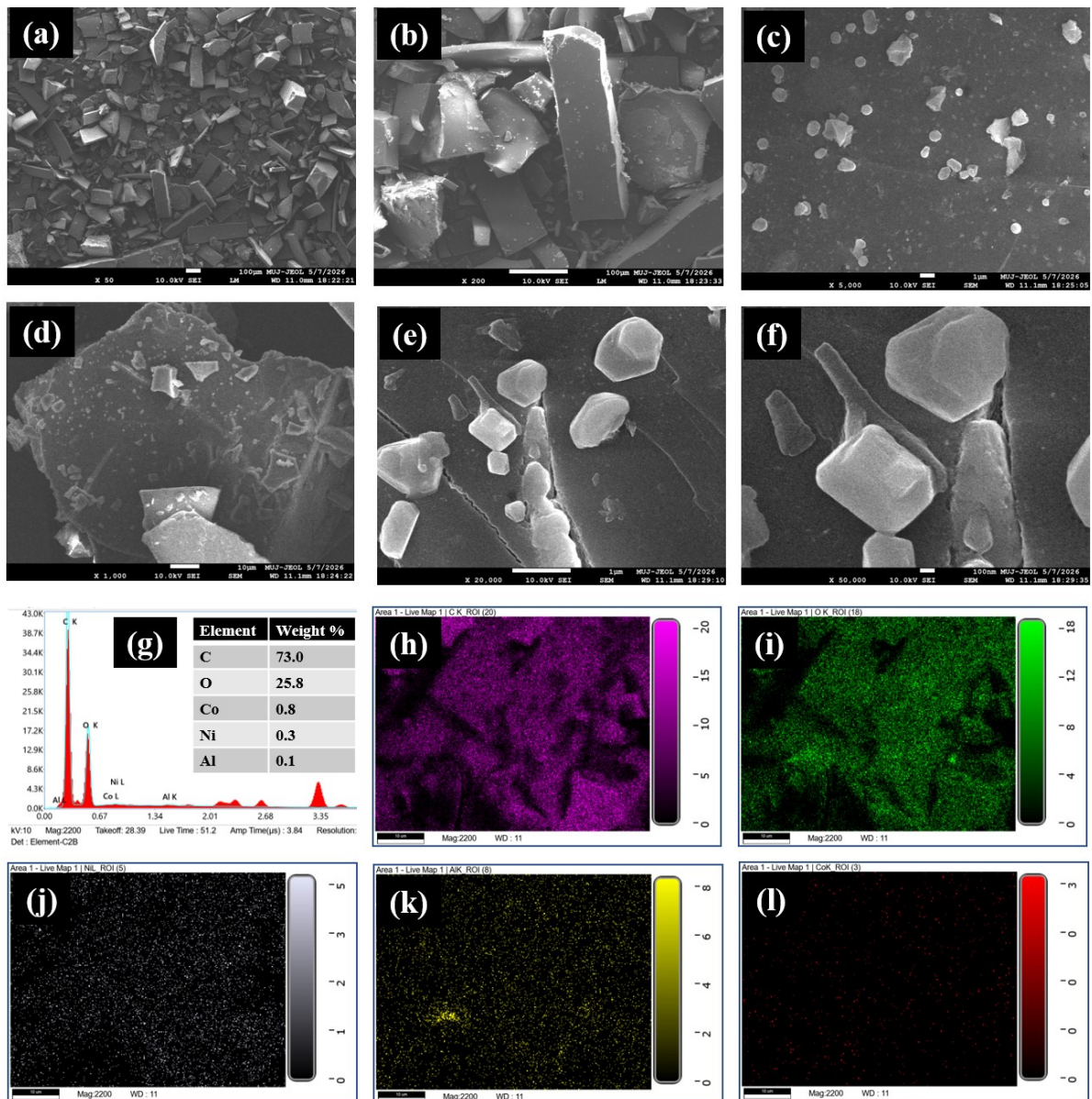


Figure S9 SEM characterization of LCO-C (a-f) SEM micrographs at different magnifications; (g) EDX spectrum and (h-l) elemental mapping of C, O, Ni, Al and Co respectively

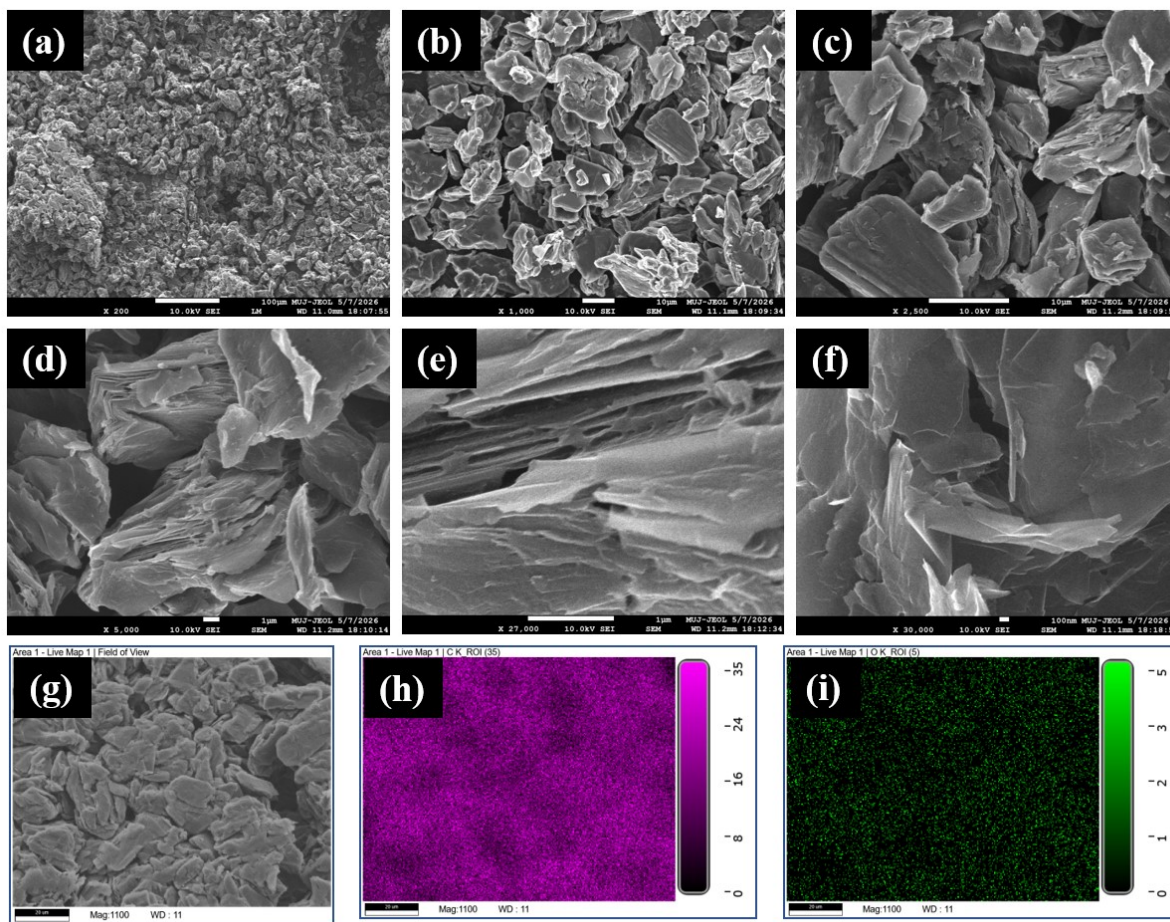


Figure S10 SEM characterization of GR-C (a-g) SEM micrographs at different magnifications and (h,i) elemental mapping of C and O respectively

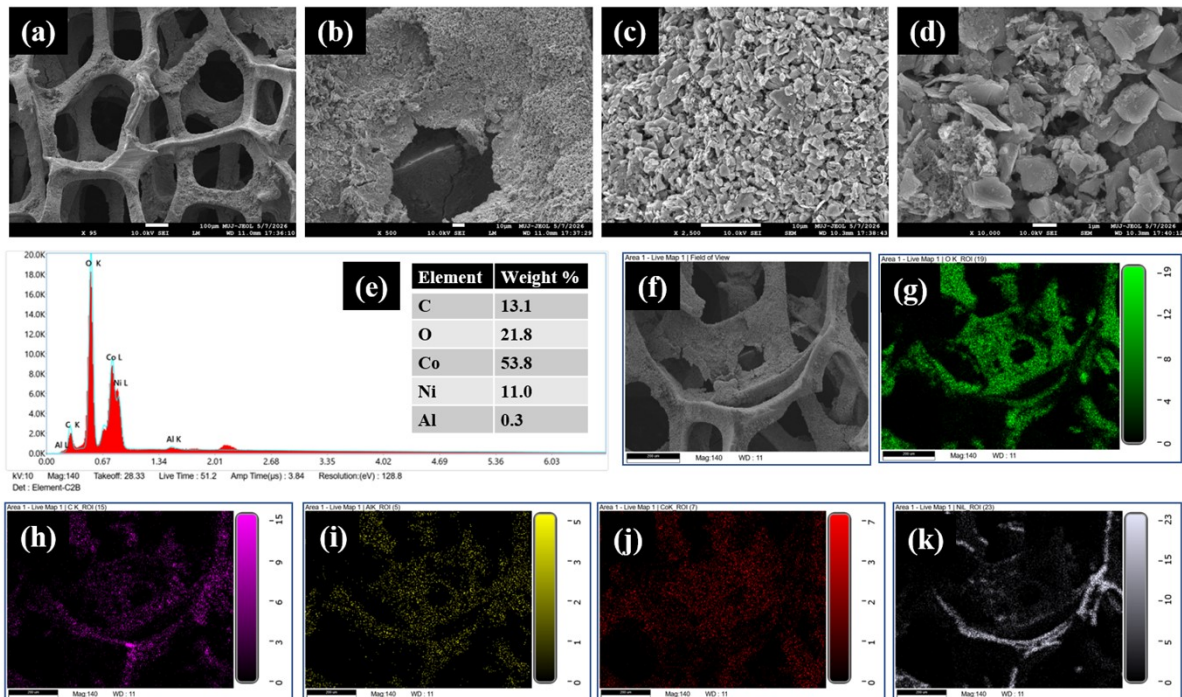


Figure S11 SEM characterization of LCO-BM_NF (a-d) SEM micrographs at different magnifications; (e) EDX spectrum; (f) SEM image selected for elemental mapping and (g-k) corresponding elemental mapping of O, C, Al, Co and Ni respectively

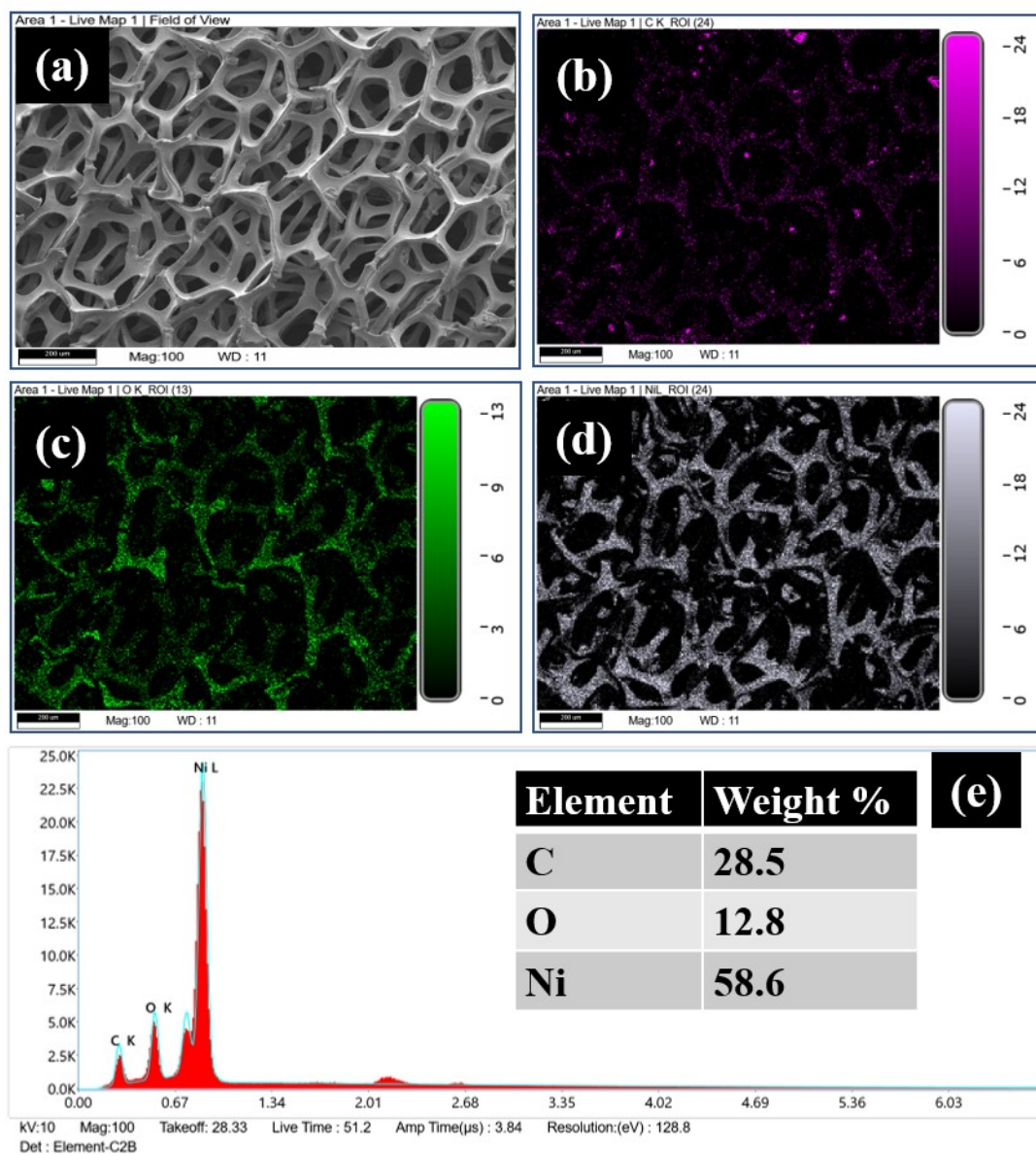


Figure S12 SEM characterization of GR-C_NF (a) SEM image selected for elemental mapping; (b-d) elemental mapping of C, O and Ni respectively and (g) EDX spectrum

Table S1: Literature survey of electrode materials recovered from spent LIBs for SC development

| Electrode | Material recovered | Recovery Method | Electrode Synthesis Methods | Specific Capacitance (F/g) | Cyclic stability (%) | Energy density (Wh/kg) | Power density (W/kg) | Ref. |
|-------------------------------------|--------------------|-----------------|-----------------------------|----------------------------|----------------------|---------------------------|-------------------------|------|
| Co ₃ O ₄ -450 | Cathode | Leaching | Calcination | 98 mF/cm ² | 100 (1000 cycles) | 0.022 mWh/cm ² | 0.84 mW/cm ² | 16 |
| LIB cathode + 10 % CNT | Cathode | Calcination | Doping | 165 | 92 (1000 cycles) | 33.4 | 3239 | 18 |
| Co ₃ O ₄ | Cathode | Leaching | Electrodeposition | 1273 | 96 (5000 cycles) | - | - | 17 |
| CNMOH | Cathode | Leaching | - | 1219 mF/cm ² | 100 (10,000 cycles) | 8.78 | 263.09 | 19 |
| Li-Ni-Mn- | Cathode | Leaching | Electrodeposition | 951 | 90 (10,000 cycles) | 30 | 377 | 20 |

| | | | | | | | | |
|-----------------|------------|----------|-------------------|------|------------------|-------|--------|----|
| Co hydroxides | | | | | | | | |
| TRGNs-Mo | Anode | - | Thermal reduction | 415 | 88 (5000 cycles) | 14.42 | 103.86 | 21 |
| S ₂ | | | | | | | | |
| GO+GQD | Anode | - | Chemical method | 2519 | 88 (1000 cycles) | 126 | - | 22 |
| Graphite@M | Black mass | Leaching | Chemical method | 270 | 80 (5000 cycles) | 0.85 | 1900 | 23 |
| nO _x | | | | | | | | |

Table S2: Electrochemical performance parameters of synthesized electrodes

| Electrode | Oxidation peak (V) | Current (mA) | CV-derived specific capacitance (F/g) | GCD-derived specific capacitance (F/g) |
|-----------|--------------------|--------------|---------------------------------------|----------------------------------------|
| LCO-S_NF | 0.37 | 65.73 | 826.46 | 557.59 |
| LCO-BS_NF | 0.48 | 55.20 | 1089.61 | 1167 |
| LCO-C_NF | 0.44 | 23.06 | 174.63 | 152.94 |
| GR-C_NF | 0.41 | 41.28 | 433.07 | 281.56 |
| GR-S_NF | 0.44 | 21.94 | 174.63 | 146.25 |
| GR-BS_NF | 0.46 | 7.31 | 73.61 | 82.11 |

Table S3: Comparison of LCO-BM_NF asymmetric with GR-C_NF with some reported Co-based two-electrode asymmetric systems

| Material | Specific capacitance (F/g) | Electrolyte used | Potential window (V) | Energy density (Wh/kg) | Power density (W/kg) | Cyclic stability (%) | Reference |
|--------------------------------------------------------------|----------------------------|------------------------------------|----------------------|------------------------|----------------------|----------------------|---------------|
| Co(OH) ₂ PC | 260 at 2 A/g | 3 M KOH | 1.5 | 76.7 | 1416.9 | 91 (5000 cycles) | ⁵⁷ |
| Co ₂ Mn ₃ O ₈ HBFC | - | 2 M KOH | 1.7 | 24.3 | 850 | 91.7 (10,000 cycles) | ⁵⁸ |
| NiCuCoO AC | 105 at 1A/g | 3 M KOH | 1.6 | 96 | 841 | 95 (5000 cycles) | ⁵⁹ |
| Co ₃ O ₄ -CQD AC | 51 at 0.5 A/g | 1 M KOH | 1.4 | 13.88 | 684.65 | 96 (5000 cycles) | ⁶⁰ |
| Co(OH) ₂ @PANI AC | 385 at 1 A/g | 6 M KOH | 1.4 | 31.2 | 1050 | 74.8 (5000 cycles) | ⁶¹ |
| MnS/Ni ₃ S ₂ @Co(OH) ₂ AC | 219.5 1 A/g | 1 M KOH | 1.5 | 68.6 | 750 | 91.2 (10,000 cycles) | ⁶² |
| Fe-CoWO ₄ AC | 82.6 at 1 A/g | 3 M KOH | 1.5 | 25.8 | 750 | 77.3 (10,000 cycles) | ⁶³ |
| CoPO AC | - | 2 M H ₂ SO ₄ | 1.4 | 24.02 | 299.21 | 99 (5000 cycles) | ⁶⁴ |
| CoMn ₄ AC | 81.8 at 0.25 A/g | 6 M KOH+PVA | 1.6 | 29.1 | 320 | 96.5 (10,000 cycles) | ⁶⁵ |
| LCO-BM_NF GR-C_NF | 16.35 at 0.5 A/g | 6 M KOH | 2 | 90.87 | 5,000 | 53 (10,000 cycles) | Current work |