

## Supporting Information

# Interfacial Superassembly of Grape-like MnO-Ni@C Frameworks for Superior Lithium Storage

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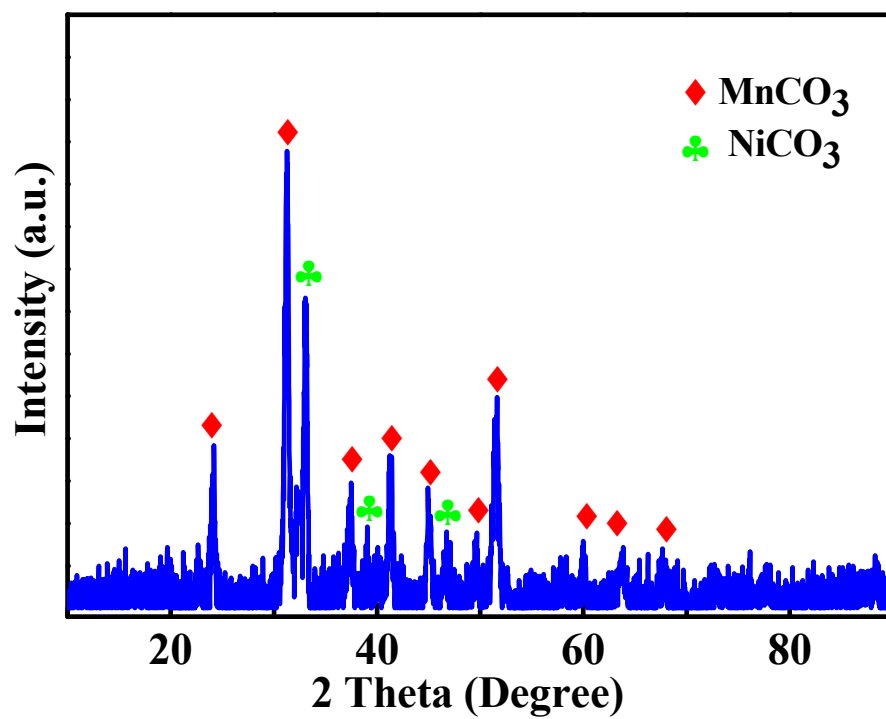
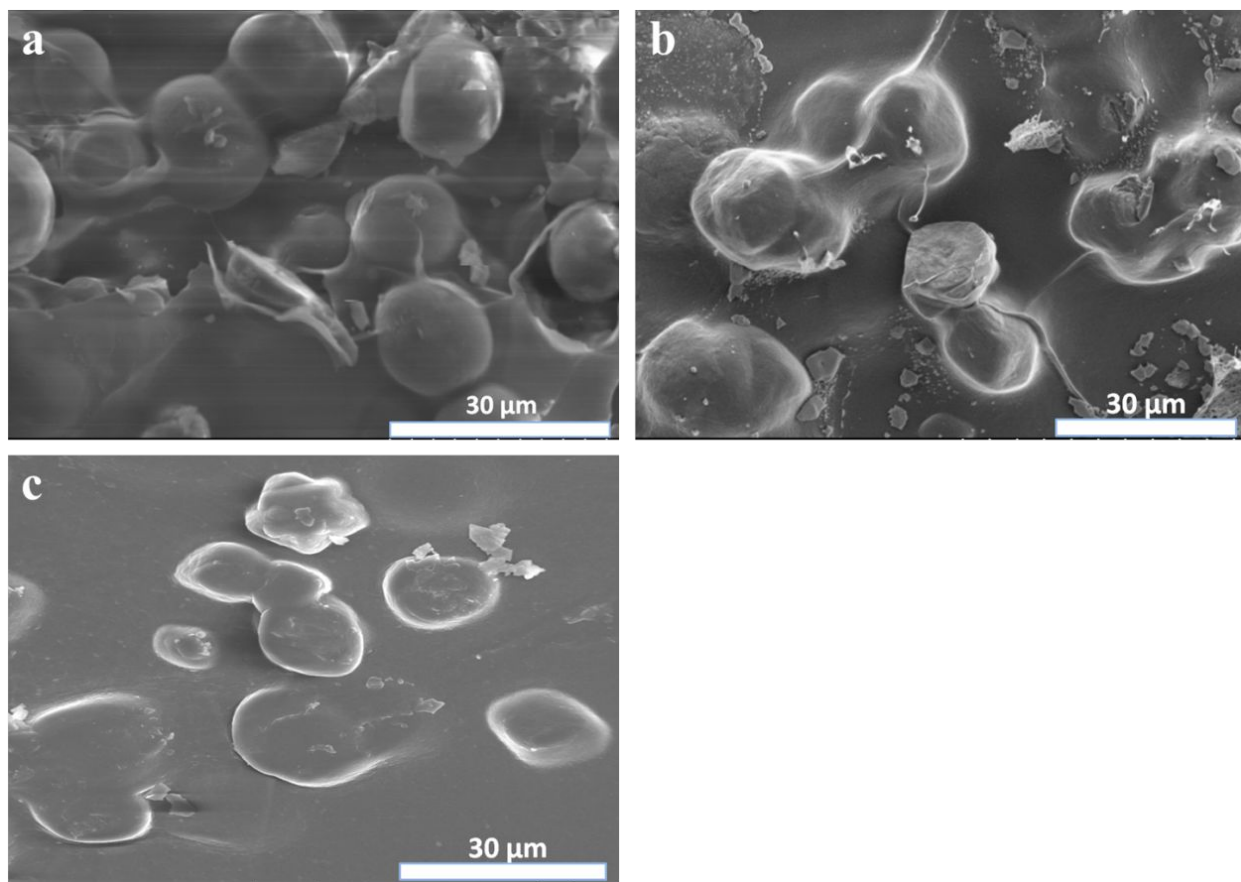
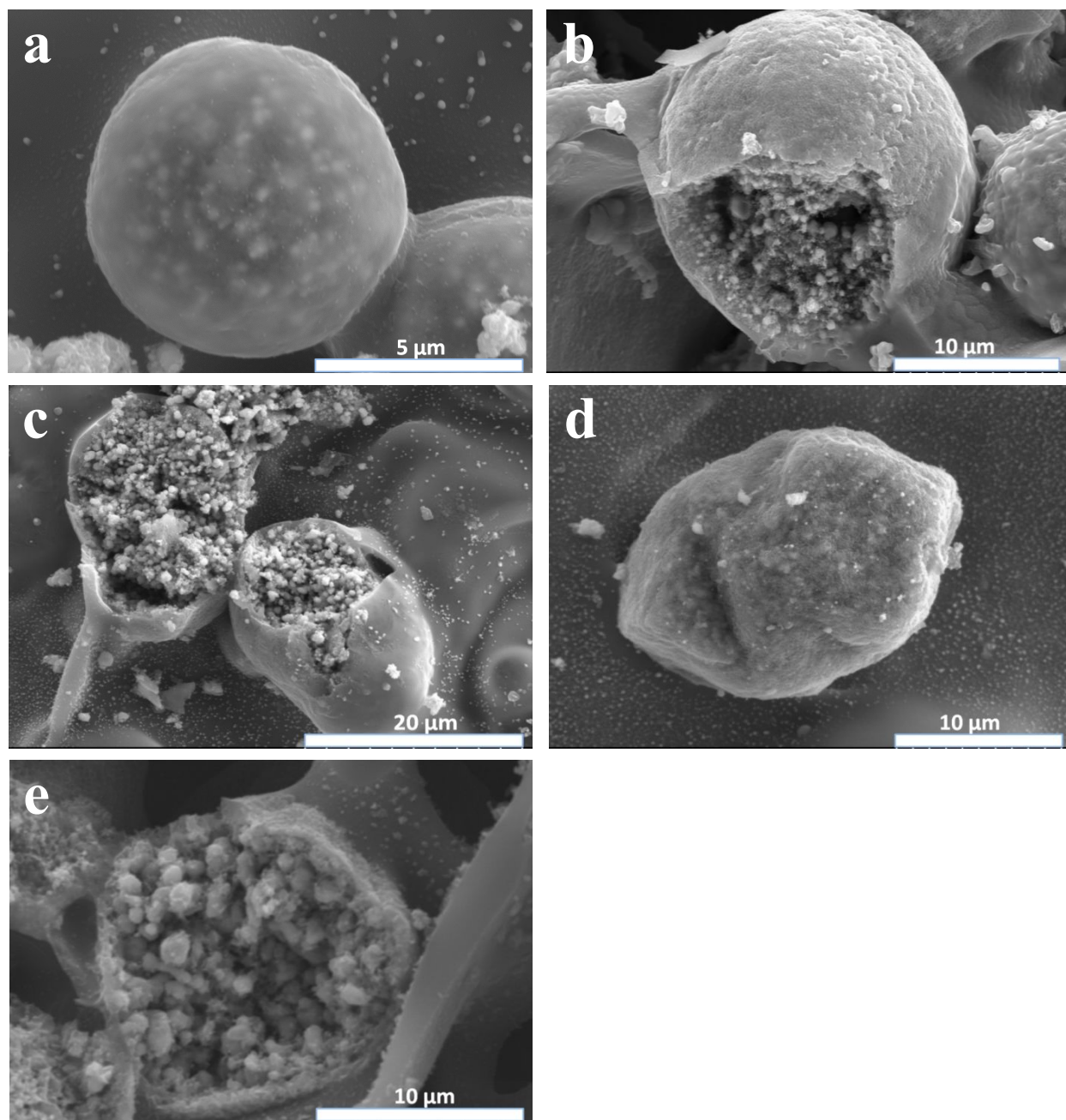


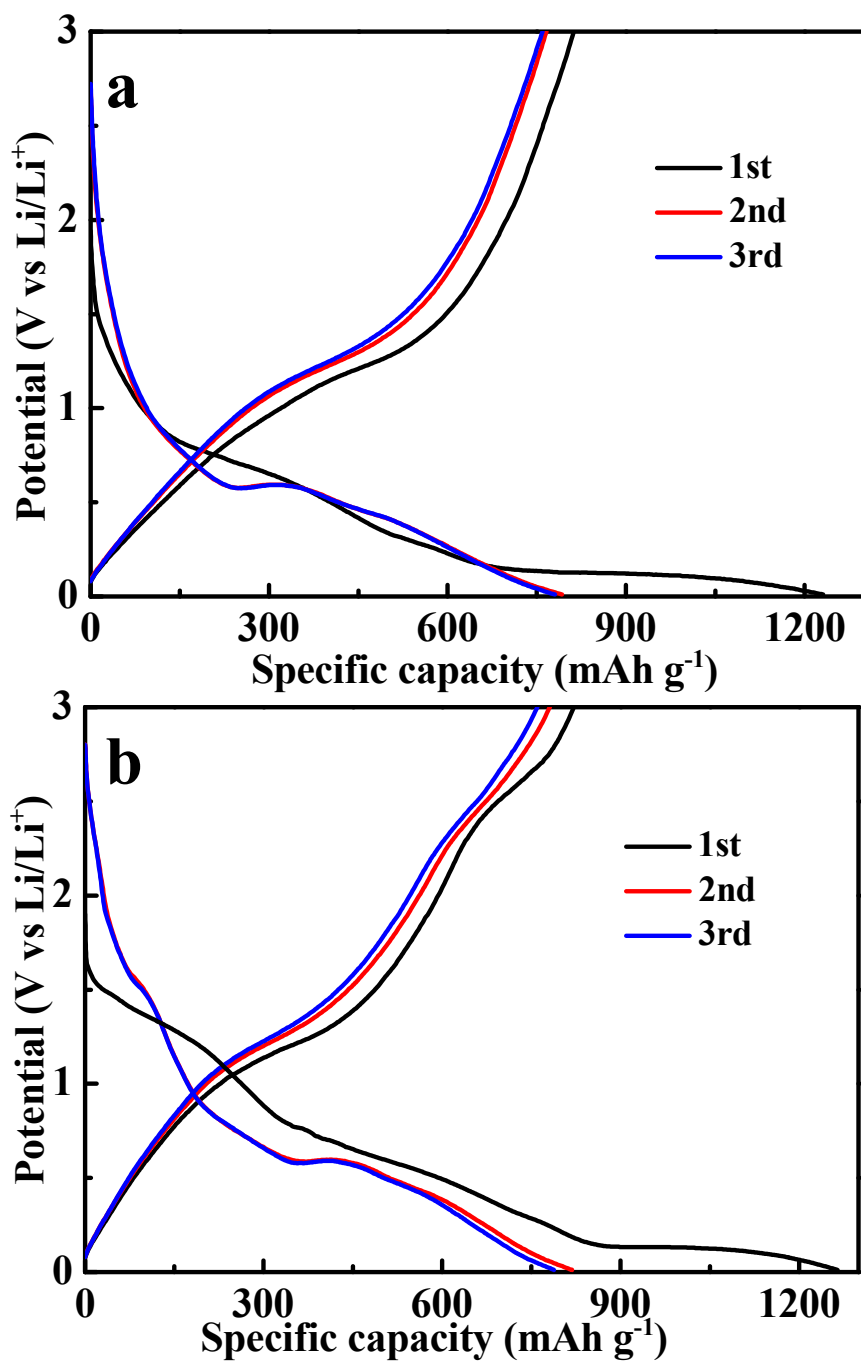
Figure S1. XRD pattern of  $\text{MCO}_3$ .



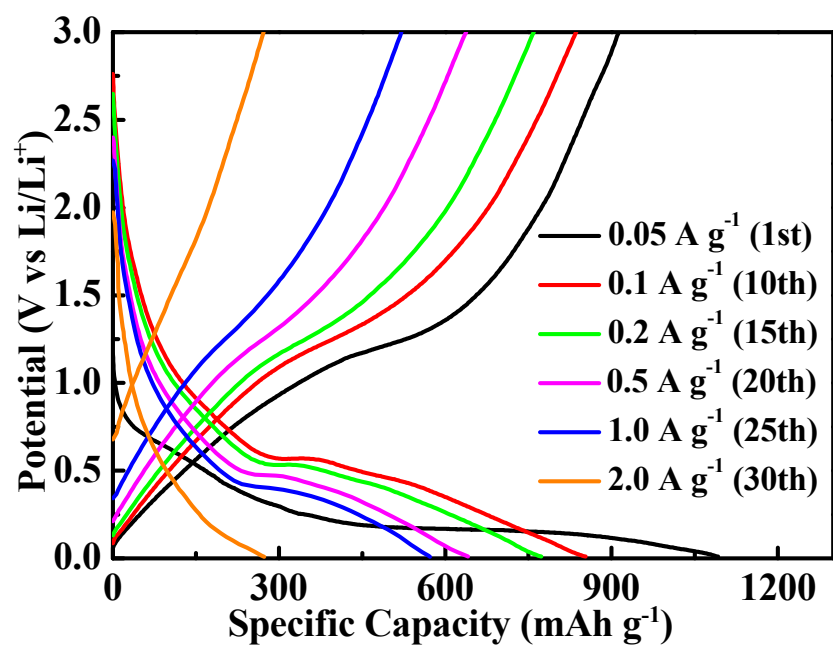
**Figure S2.** SEM images of  $\text{MCO}_3/\text{ager}$  gel matrix precursor: 5% (a); 10% (b) and 15% Ni (c).



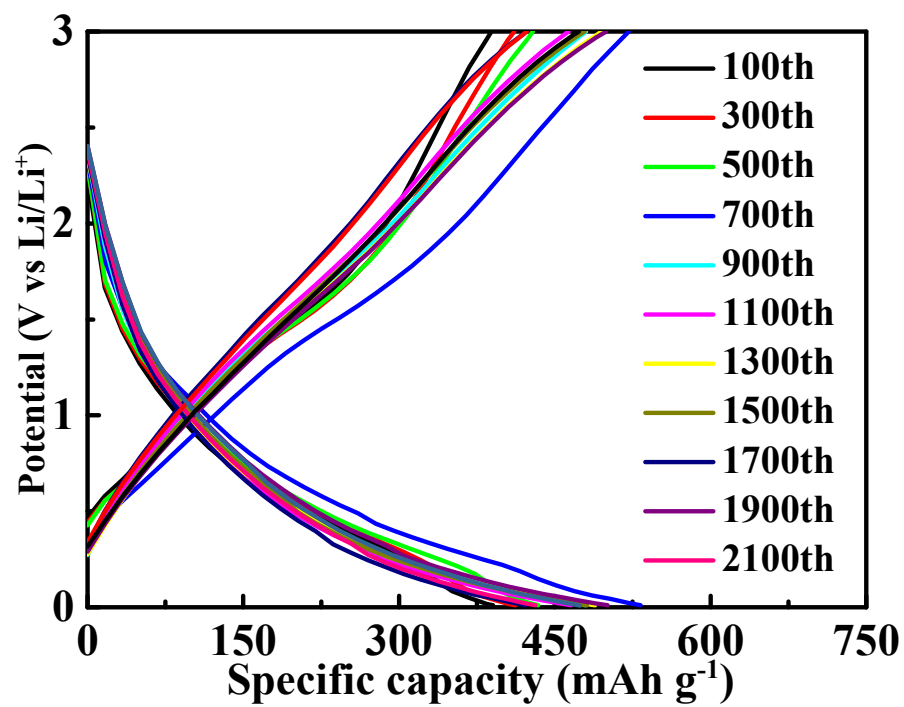
**Figure S3.** SEM images of MnO-Ni5@C (a, b), MnO-Ni10 (c) and MnO-Ni15@C (d, e).



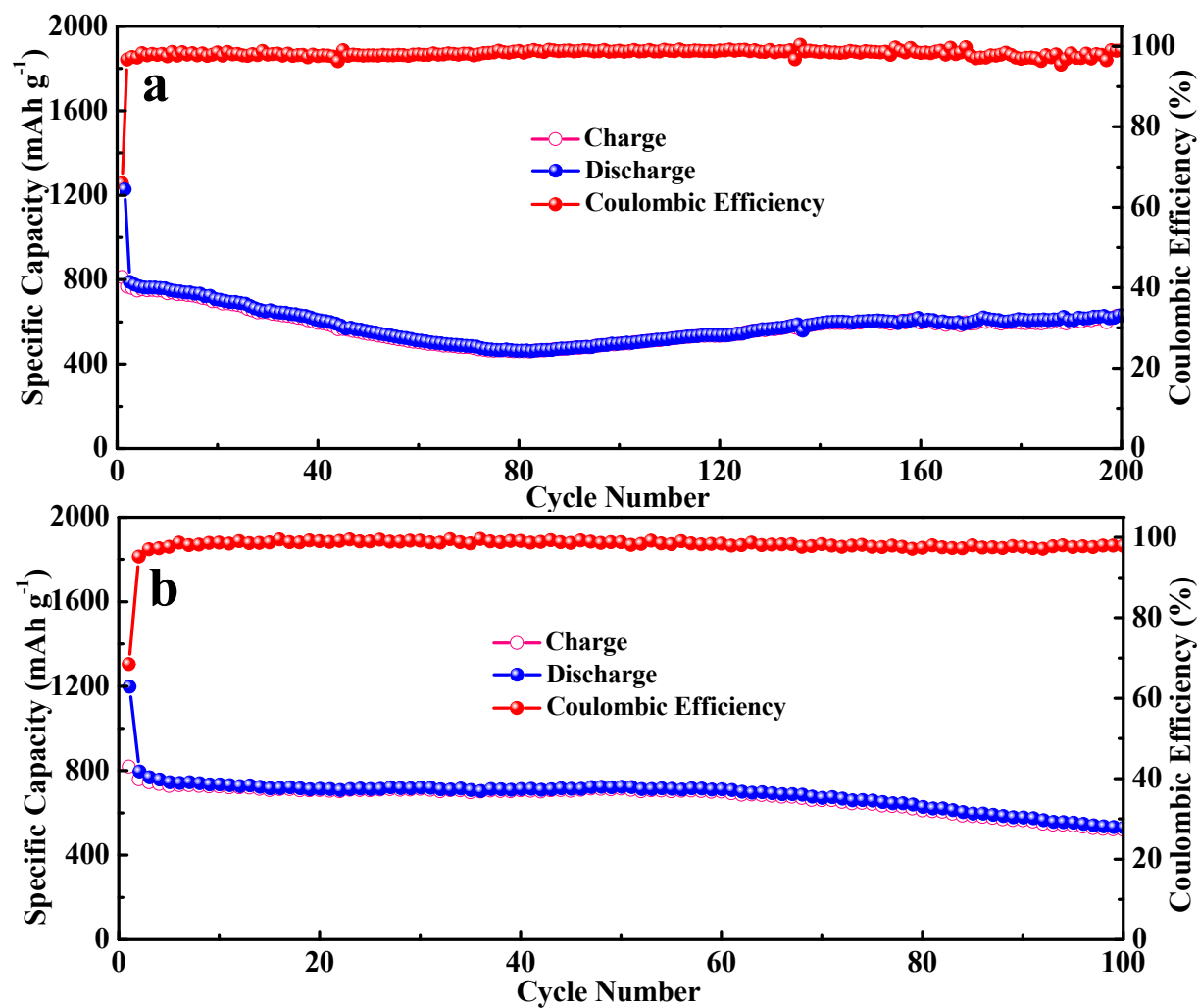
**Figure S4.** Charge-discharge profiles of MnO-Ni<sub>5</sub>@C (a) and MnO-Ni<sub>15</sub>@C (b) electrodes at the current density of 0.1 A g<sup>-1</sup>.



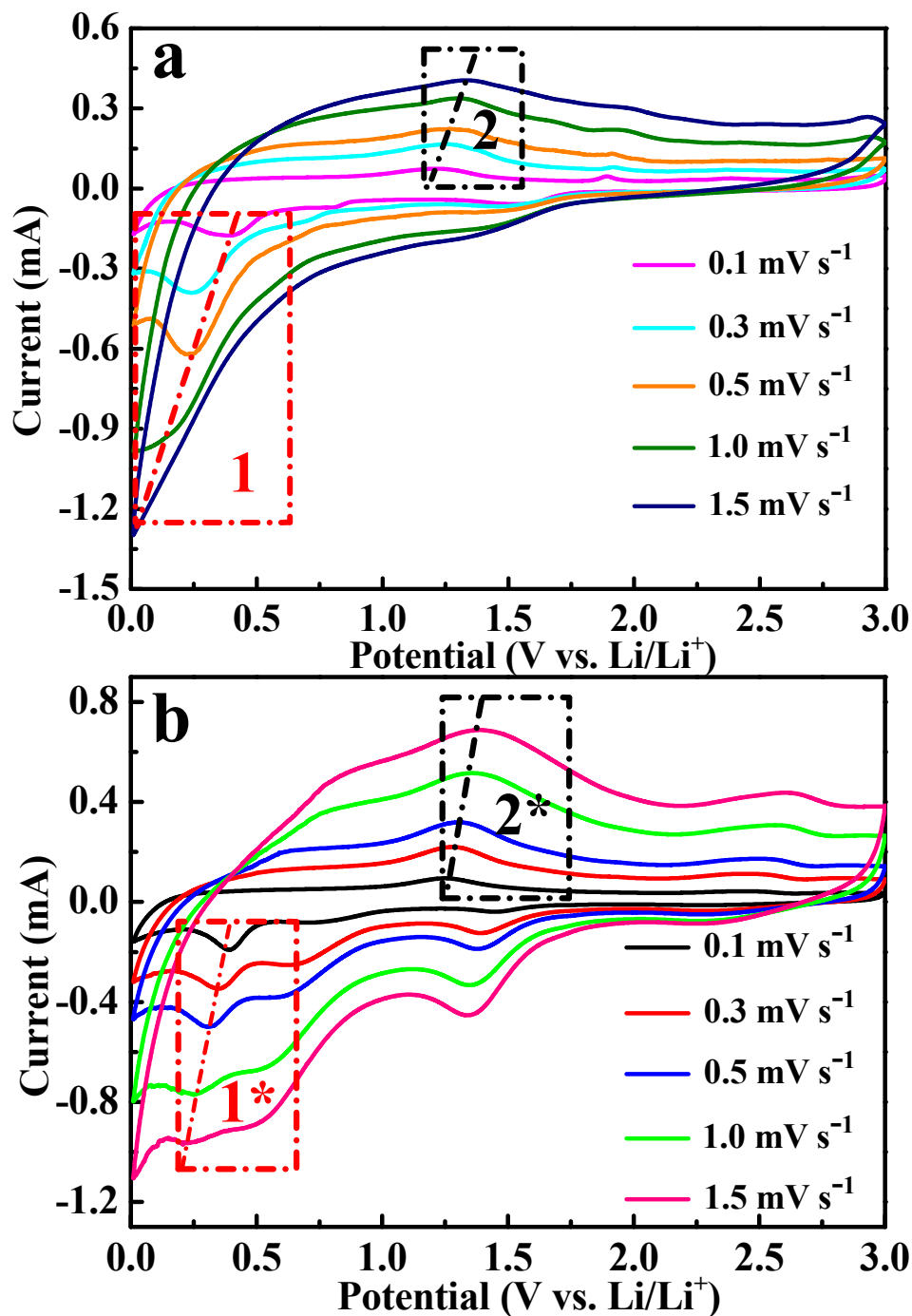
**Figure S5.** Charge-discharge profiles of MnO-Ni10@C electrodes at different current densities.



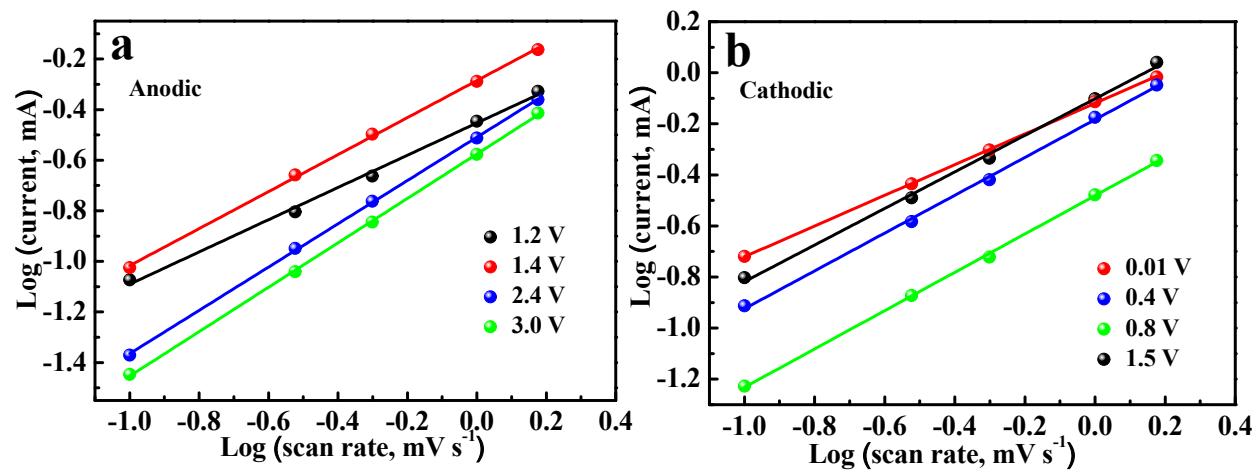
**Figure S6.** The corresponding selected charge-discharge profiles of MnO-Ni10@C at 1.0 A g<sup>-1</sup> for a ultralong cycle life of 2100 cycles.



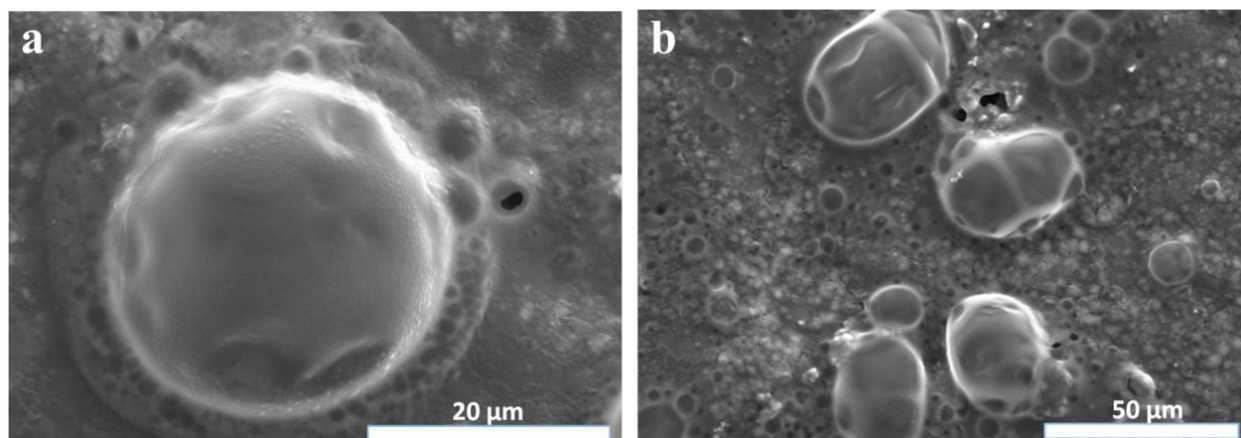
**Figure S7.** Cycling performance and coulombic efficiency of MnO-Ni5@C (a) and MnO-Ni15@C (b) electrodes at current density of 0.1 A g<sup>-1</sup>.



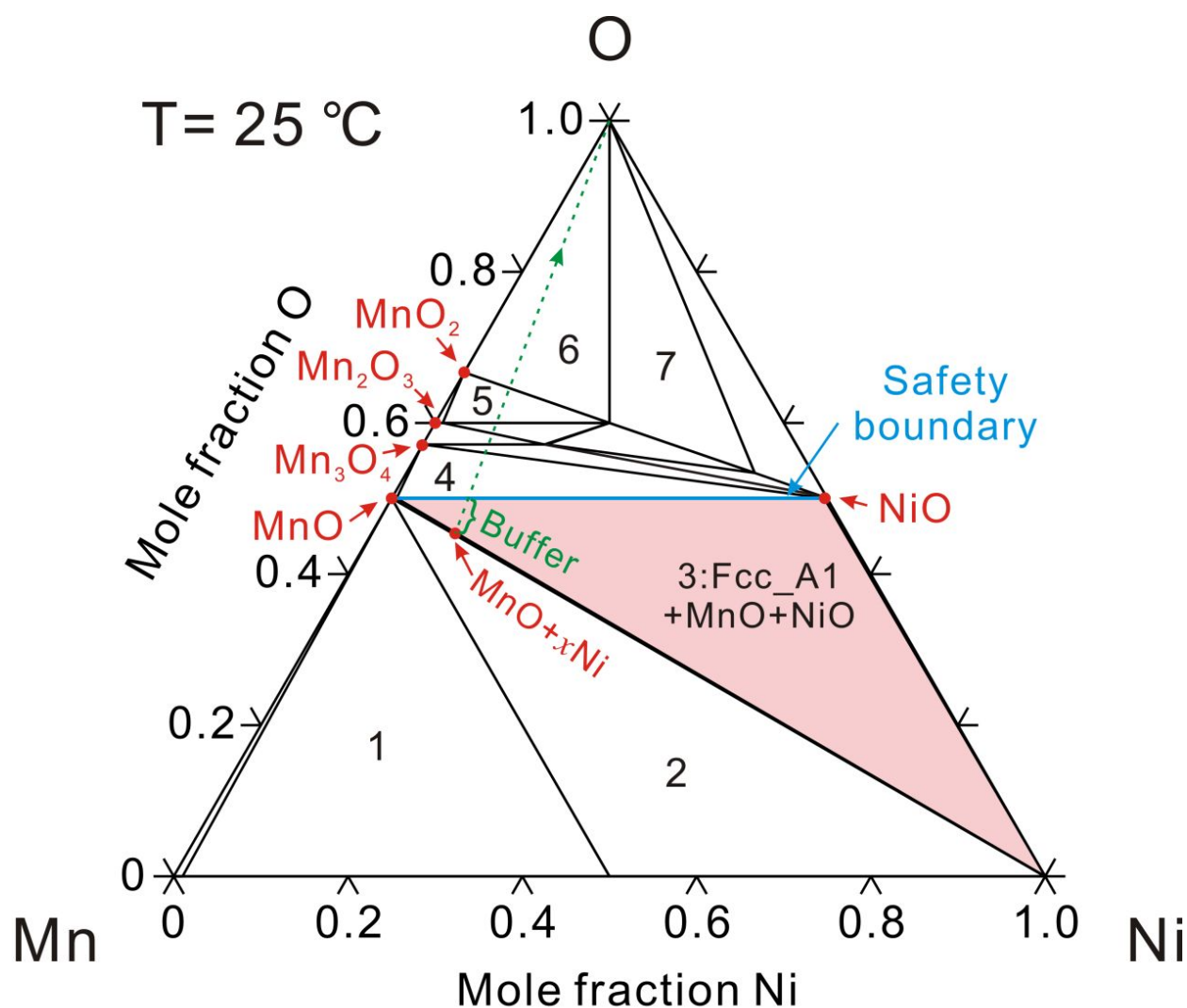
**Figure S8.** CV curves at differet scan rates from 0.1-1.5 mV s<sup>-1</sup> of MnO-NiO@C (a) and MnO-Ni10@C (b) electrodes.



**Figure S9.** Current responses plotted against different scan rates of MnO-Ni10@C electrodes at different potentials for anodic scans (a) and cathodic scans (b).



**Figure S10.** SEM images of MnO-Ni<sub>5</sub>@C (a) and MnO-Ni<sub>15</sub>@C (b) electrodes after 200 and 100 cycles, respectively.



**Figure S11.** The calculated room temperature phase diagram of Ni-Mn-O system. The dashed line is the oxidation direction of the  $\text{MnO} + x\text{Ni}$  alloys.

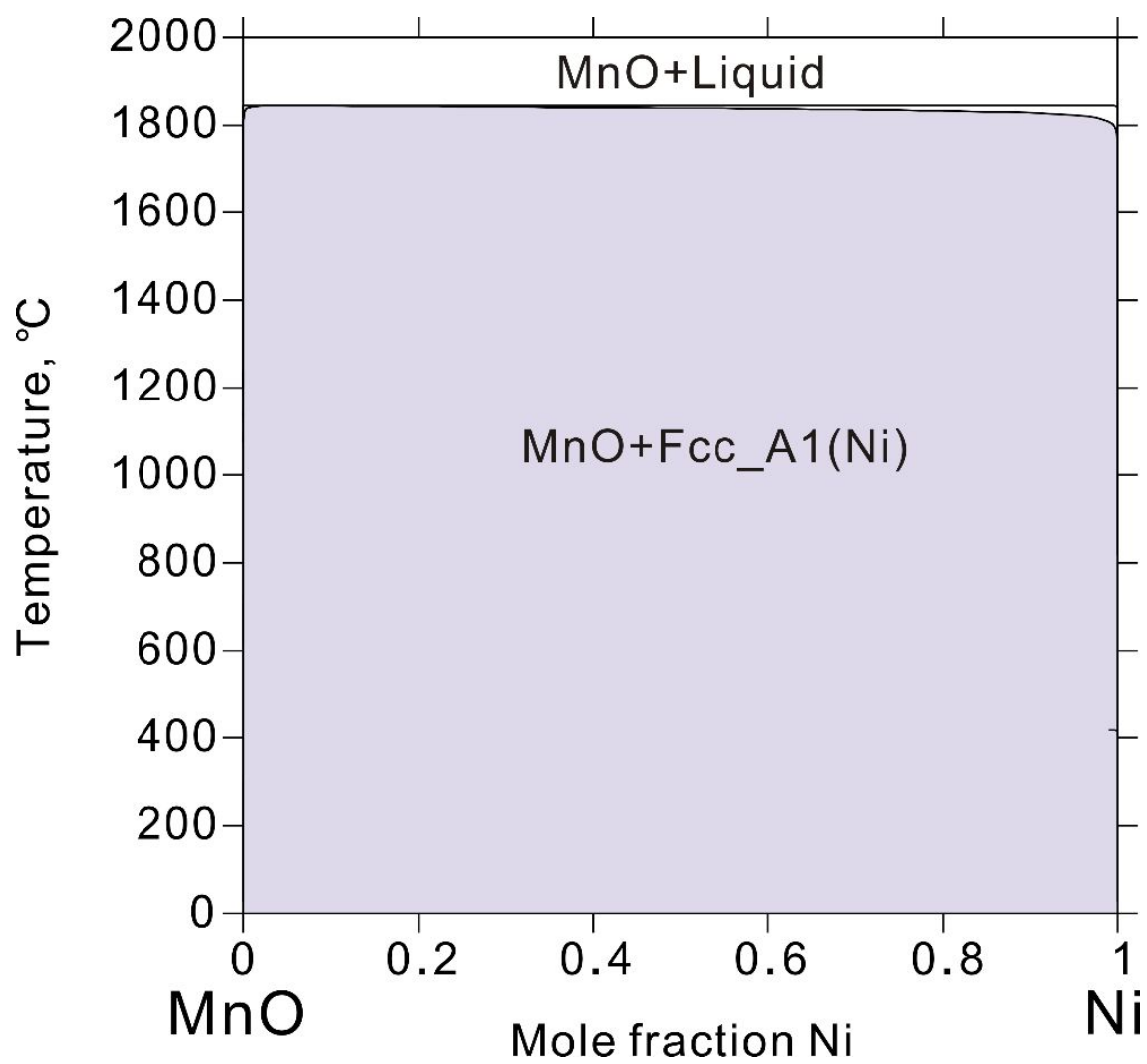


Figure S12. Calculated MnO-Ni vertical section of the Ni-Mn-O system.

**Table S1.** Comparisons of the electrochemical performance of the transition metal oxides electrodes.

| MnO-based Materials                | Current Density<br>[A g <sup>-1</sup> ] | Cycle Numbers | Initial Value<br>(the 2 <sup>th</sup> cycle)<br>[mAh g <sup>-1</sup> ] | Crest Value<br>[mAh g <sup>-1</sup> ] | Retention of Discharge Capacity | References |
|------------------------------------|-----------------------------------------|---------------|------------------------------------------------------------------------|---------------------------------------|---------------------------------|------------|
| MnO@Mn <sub>3</sub> O <sub>4</sub> | 0.2                                     | 300           | 748                                                                    | 1300                                  | 173.79%                         | 1          |
| MnO/C-N                            | 0.5                                     | 500           | 582                                                                    | 783                                   | 134.53%                         | 2          |
| CNT/Co <sub>3</sub> O <sub>4</sub> | 0.1                                     | 100           | 900                                                                    | 1200                                  | 133.33%                         | 3          |
| Co <sub>3</sub> O <sub>4</sub>     | 1.0                                     | 1000          | 762                                                                    | 1910                                  | 250.65%                         | 4          |
| MnO@Graphene                       | 0.2                                     | 100           | 805                                                                    | 1202                                  | 149.32%                         | 5          |
| RGO-MnO-RGO                        | 2.0                                     | 500           | 600                                                                    | 1269.2                                | 211.53%                         | 6          |
| MnO/carbon matrix                  | 0.1                                     | 100           | 760                                                                    | 952                                   | 125.26%                         | 7          |
| MnO/C                              | 1.0                                     | 1000          | 890                                                                    | 1212                                  | 136.18%                         | 8          |
| MnO/N-Doped C                      | 0.1                                     | 200           | 1231.1                                                                 | 1699                                  | 138%                            | 9          |
| MnO /Carbon                        | 0.5                                     | 170           | 420                                                                    | 1467.                                 | 349.29%                         | 10         |
| MnO Nanoparticles                  | 5.0                                     | 5000          | 825                                                                    | 939                                   | 113.82%                         | 11         |
| MnO/C nanowires                    | 0.1                                     | 100           | 635                                                                    | 832                                   | 131.02%                         | 12         |
| MnO on Graphene                    | 0.2                                     | 150           | 890.7                                                                  | 2014.1                                | 226.13%                         | 13         |
| Nano-MnO/C                         | 0.1                                     | 200           | 895                                                                    | 1082                                  | 120.89%                         | 14         |
| MnO@C Nanowires                    | 0.5                                     | 200           | 650                                                                    | 801                                   | 123.23%                         | 15         |
| MnO/Carbon                         | 0.5                                     | 300           | 735                                                                    | 1453                                  | 197.69%                         | 16         |
| MnO@C                              | 0.1                                     | 270           | 673                                                                    | 1450.5                                | 215.53%                         | 17         |
| MnO/N-doped C                      | 0.5                                     | 100           | 795                                                                    | 982                                   | 123.52%                         | 18         |
| <b>This work</b>                   | 0.1                                     | 200           | <b>820</b>                                                             | <b>832</b>                            | <b>101.46%</b>                  |            |

## Notes and references

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