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Enhanced charge storage capacity and high rate capabilities of Ni_2Co-layered double hydroxides/expanded graphite composites as an anode for Li-ion batteries

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Computational Details and Modelling:

All *ab initio* calculations were performed with the Vienna *Ab initio* Simulation Package (VASP 5.4.4).[1-4] We used the projector augmented wave (PAW) method [5, 6] with the generalized gradient approximation based on the Perdew-Burke-Ernzerhof (PBE) [7] functional including the Hubbard *U* correction (GGA+*U*). [8] Plane-wave cutoff energy of 500 eV was used. Lattice constants and internal atomic positions were fully optimized until the residual forces were less than 0.04 eV/ Å. The vacuum slab space of a unit cell in the z-direction was set to 20 Å to avoid interactions between layers. To satisfy the experimental transition metal atomic ratio for Ni_2Co-LDH, a model was constructed using a 3x3 supercell as $Ni_6Co_3O_{18}H_{18}$ containing 9 transition metal sites. The Brillouin zone was performed with a k-point grid of 3 x 3 x 1 based on the Monkhorst-Pack scheme[9] using a k-point mesh with an interval of 0.05 $Å^{-1}$.

Theoretical capacities calculation:

To quantitatively distinguish the lithium storage ability of $Ni_{2/3}Co_{1/3}(OH)_2$, we calculated theoretical specific capacity by using the following equation:

$$Q_{theoretical} = \frac{(nF) \times 1000}{(3600 \times MW)} \text{ mAh/g}$$

where *n* is the number of charge carriers, *F* is the Faraday constant (96485 C/mol) and *MW* is the molecular weight of the $Ni_{2/3}Co_{1/3}(OH)_2$ (92.7871 g/mol). Finally, to use capacity units in mAh/g, multiply by 1000.

I. $Ni_{2/3}Co_{1/3}(OH)_2 + Li^+ + e^- \rightarrow Ni_{2/3}Co_{1/3}OH + LiOH$

$$Q_{theoretical} = \frac{1 \times 96485 \times 1000}{(3600 \times 92.7871)} = 289 \text{ mAh/g}$$

II. $Ni_{2/3}Co_{1/3}(OH)_2 + 2Li^+ + 2e^- \rightarrow Ni_{2/3}Co_{1/3} + 2LiOH$

$$Q_{theoretical} = \frac{2 \times 96485 \times 1000}{(3600 \times 92.7871)} = 578 \text{ mAh/g}$$

III. $Ni_{2/3}Co_{1/3}(OH)_2 + 4Li^+ + 4e^- \rightarrow Ni_{2/3}Co_{1/3} + LiOH + LiH + Li_2O$

$$Q_{theoretical} = \frac{4 \times 96485 \times 1000}{(3600 \times 92.7871)} = 1155 \text{ mAh/g}$$

IV. $Ni_{2/3}Co_{1/3}(OH)_2 + 6Li^+ + 6e^- \rightarrow Ni_{2/3}Co_{1/3} + 2LiH + 2Li_2O$

$$Q_{theoretical} = \frac{6 \times 96485 \times 1000}{(3600 \times 92.7871)} = 1733 \text{ mAh/g}$$

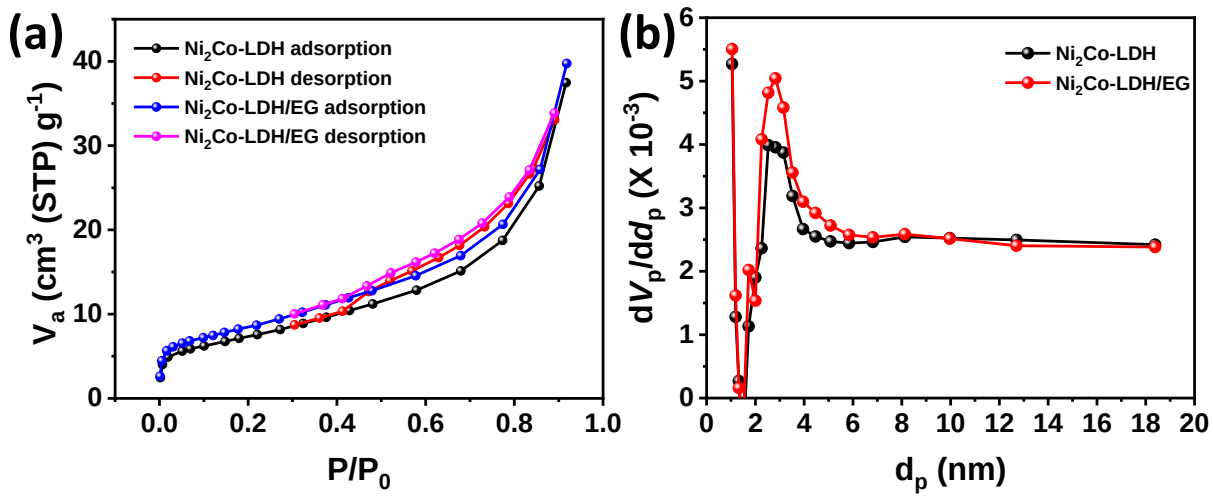

Fig. S1. (a) Adsorption-desorption isotherm, (b) BJH pore size distribution of Ni_2Co-LDH and Ni_2Co-LDH/EG composite

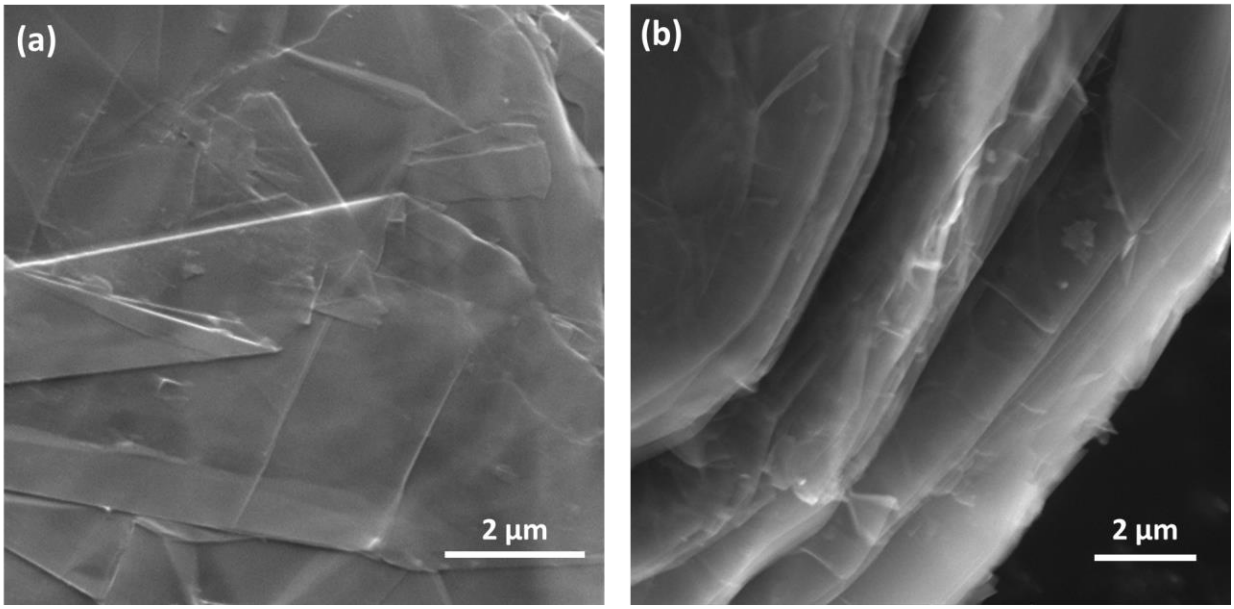

Fig. S2. FESEM image of EG (a) in-plane and (b) out of plane

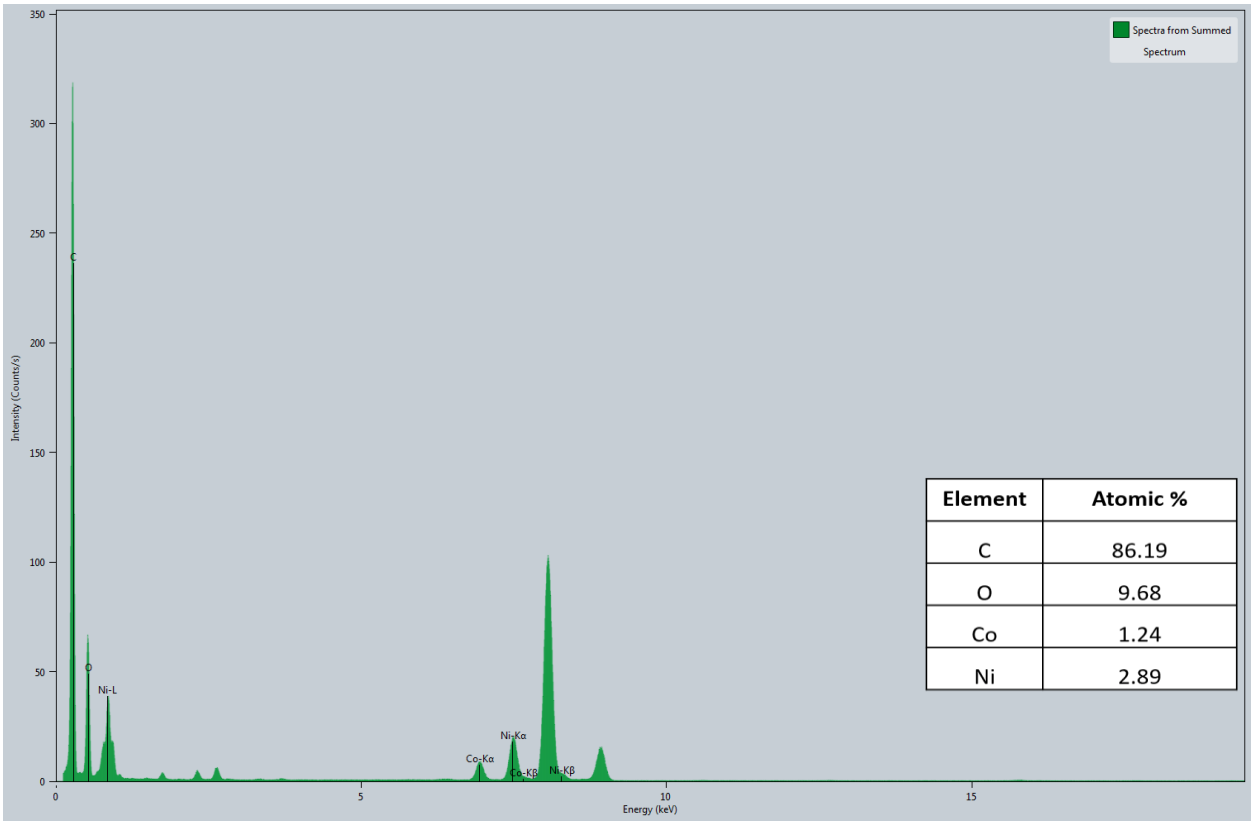

Fig. S3. Energy dispersive X-ray spectroscopy (EDS) of Ni_2Co-LDH/EG composites.

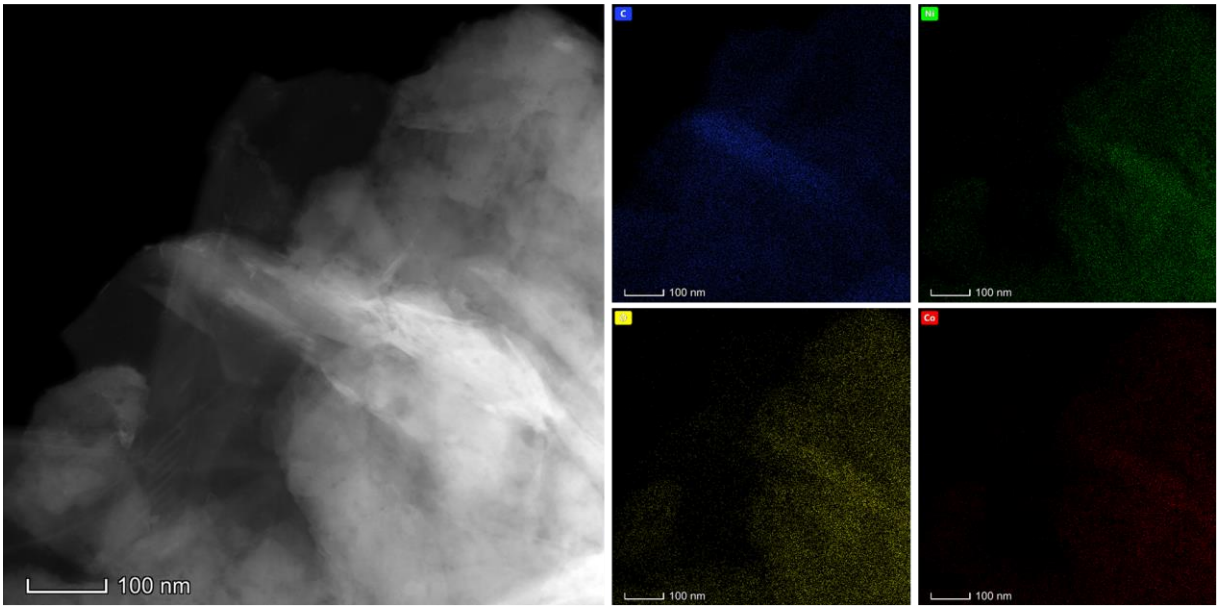

Fig. S4. Elemental mapping using HAADF-STEM of Ni_2Co-LDH/EG composites.

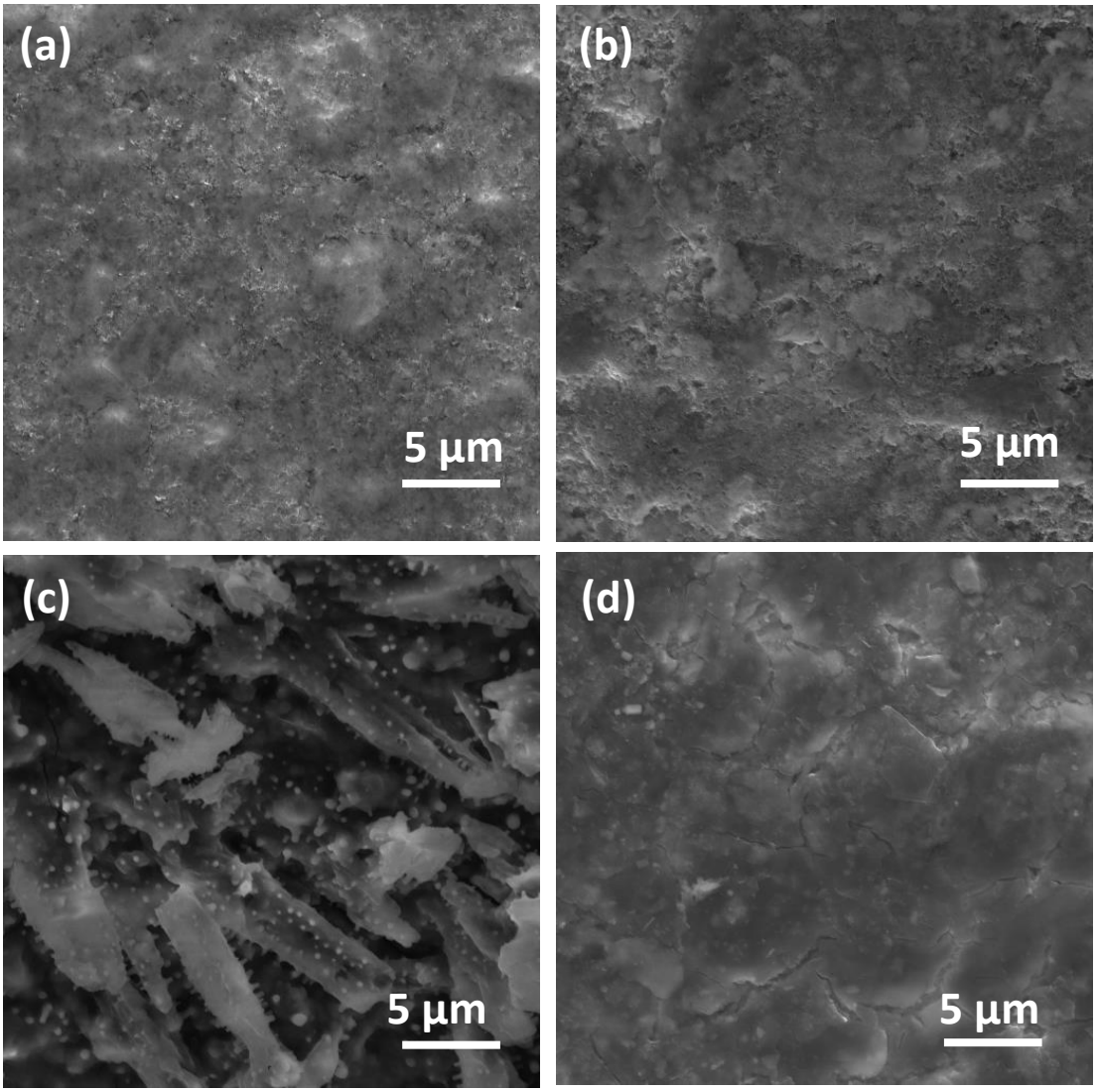

Fig. S5. FESEM image before cycling of (a) Ni_2Co-LDH, (b) Ni_2Co-LDH/EG composites films, and after cycling for 20 cycles of (c) Ni_2Co-LDH, (d) Ni_2Co-LDH/EG composites films.

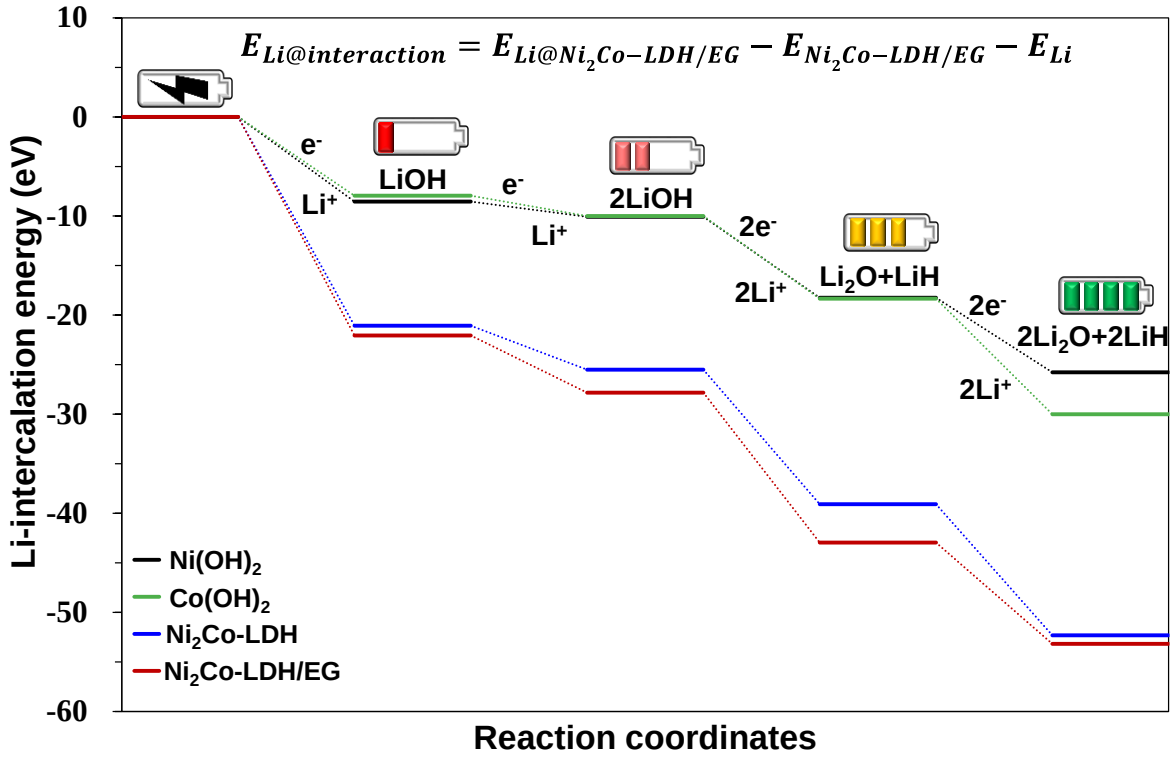

Fig. S6. Schematic representation of the Li interaction energy according to the reaction coordinates (up to step4) of $Ni(OH)_2$, $Co(OH)_2$, Ni_2Co-LDH, and Ni_2Co-LDH/EG

References

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