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Electronic Supplementary Information (ESI)

Fullerene as an efficient hybridization matrix for exploring high-performance layered-double-hydroxide-based electrodes

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Fig. S1. Colloidal suspensions of exfoliated Ni–Fe-LDH nanosheet (NS) and C_{60}.

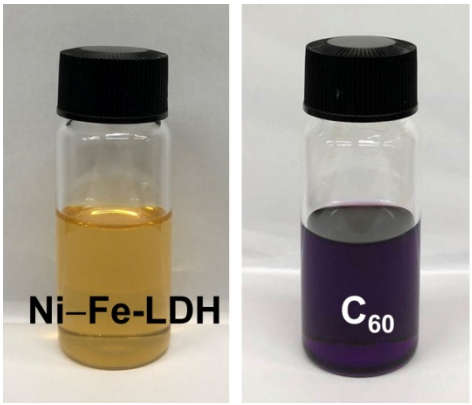


Table S1. Lattice parameters of C_{60} and **CNF** nanohybrids.

Material	a = b = c (Å)	RMS error
C_{60}	14.202	5.07E-05
CNF3	14.207	1.07E-04
CNF4	14.208	7.22E-05
CNF5	14.205	1.42E-04

Fig. S2. Zeta potential distribution of exfoliated Ni–Fe-LDH NS and C_{60}

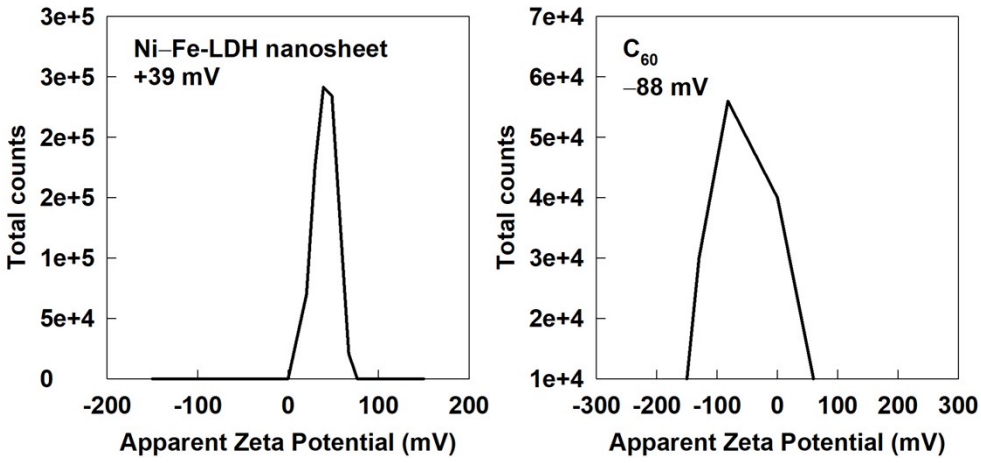


Fig. S3. Micro-Raman spectra of the **CNF** nanohybrid, Ni–Fe-LDH, and C_{60}.

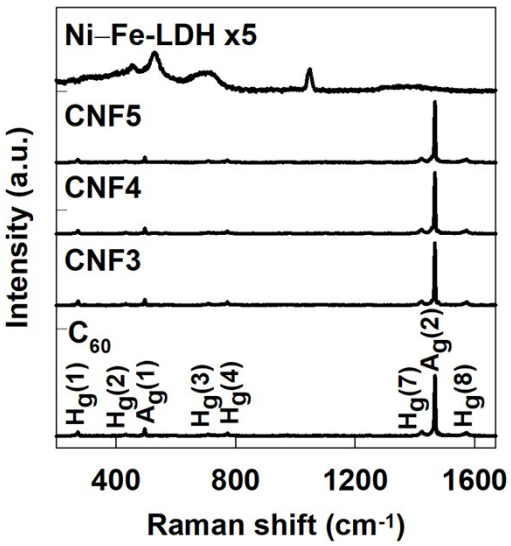


Fig. S4. Transmission electron microscopy (TEM) images of exfoliated Ni–Fe-LDH NS and C_{60}.

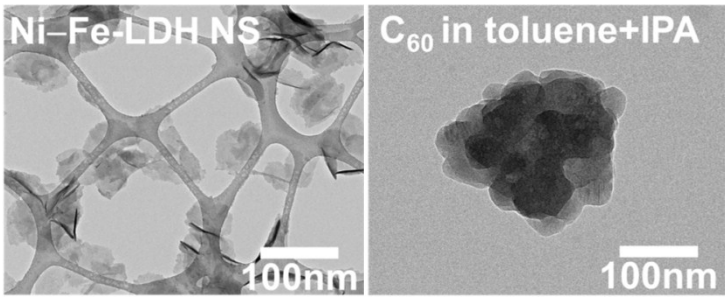


Fig. S5. Field emission-scanning electron microscopy (FE-SEM) images of **CNF3**, **CNF4**, and **CNF5** with different magnifications.

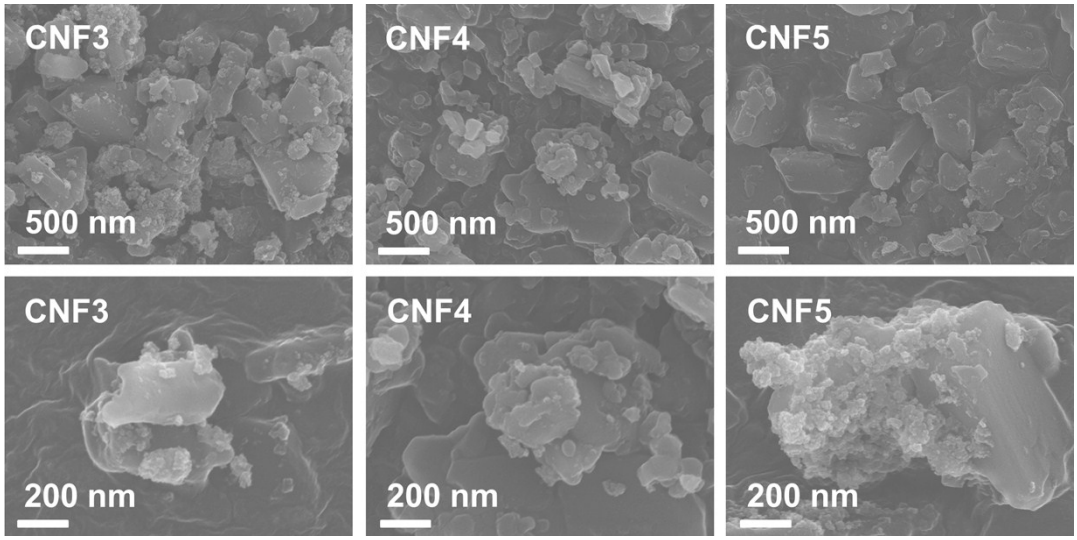


Fig. S6. TEM images of **CNF3** and **CNF5** nanohybrids.

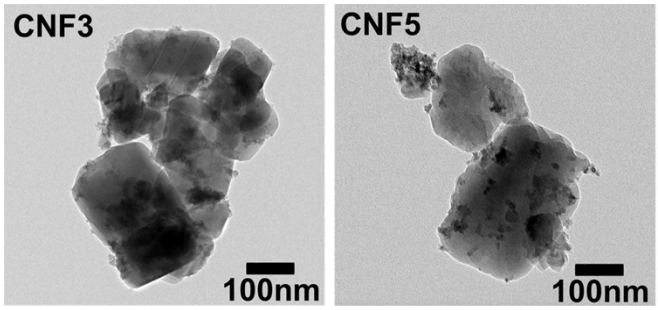


Table S2. Relative concentrations of Ni and Fe species in the present materials determined from Ni $2p_{3/2}$ and Fe $2p_{3/2}$ X-ray photoelectron spectroscopy (XPS) data.

Material	Ni^{2+}	Ni^{3+}	Fe^{2+}	Fe^{3+}
Ni–Fe-LDH	84.5 %	16.5 %	34.1 %	65.9 %
CNF3	67.2 %	32.8 %	30.0 %	70.0 %
CNF4	70.2 %	29.8 %	32.4 %	67.6 %
CNF5	71.3 %	28.7 %	33.6 %	66.4 %

Fig. S7. C 1s XPS spectra of C_{60} and **CNF** nanohybrids.

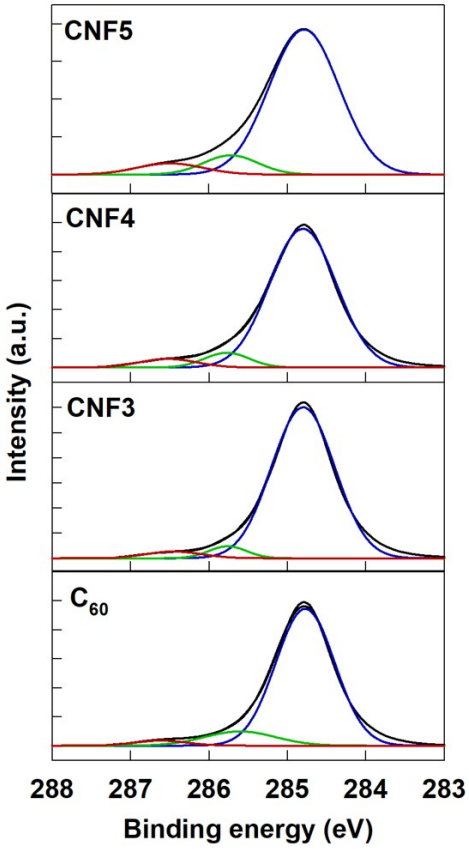


Fig. S8. Cyclic voltammetry (CV) curves of **CNF3** and **CNF5**.

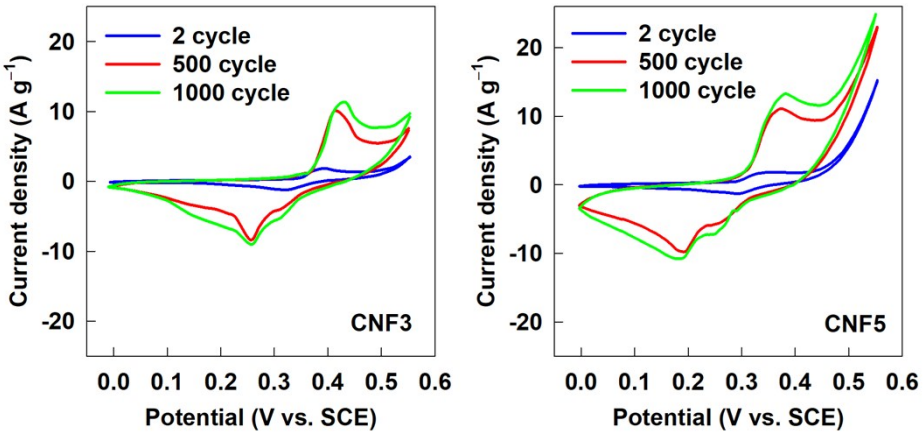


Fig. S9. Specific capacitance plots of Ni–Fe-LDH, C_{60}, and **CNF4** at a constant current density of 1 A g^{-1} .

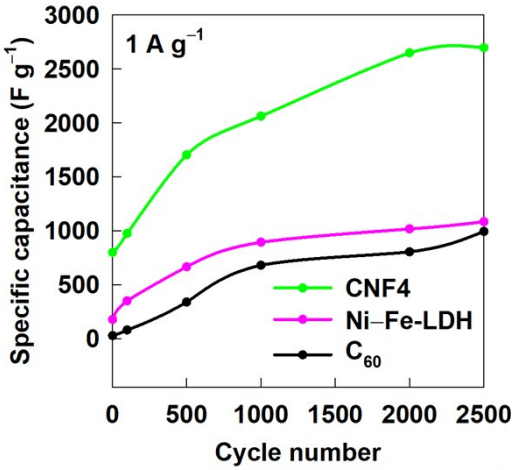


Table S3. Physical properties and specific capacitances of Ni–Fe-LDH-based electrode materials in the previous reports and current work.

Material	Specific capacitance	Method	Current density	Potential window	Electrolyte	Ref
Ni–Fe-LDH/RGO/carbon nanofiber	1330.2 F g^{-1}	CD	1 A g^{-1}	0–0.44 V	6 M KOH	1
CoNiFe-LDH/ carbon nanofiber	1203 F g^{-1}	CD	1 A g^{-1}	0–0.4 V	6 M KOH	2
N,S-codoped rGO/WSe_2/Ni–Fe-LDH	125.6 F g^{-1}	CD	1 A g^{-1}	0–0.4 V	1 M KOH	3
Ultrafine Ni–Fe-LDH nanosheet@rGO	2715 F g^{-1}	CD	3 A g^{-1}	0–1.6 V	1 M KOH	4
Sulfidation of Ni–Fe-LDH	992 mF cm^{-2}	CD	2 mA cm^{-2}	−1–0 V	1 M KOH	5
Ni–Fe-LDH/MnO_2	1127 F g^{-1}	CD	1 A g^{-1}	0–0.5 V	6 M KOH	6
Carbon cloth/$NiFe_2O_4$	922.6 F g^{-1}	CD	2 mA cm^{-2}	0–1.0 V	6 M KOH	7
Ni–Fe LDH/graphene hybrid aerogel	1196 F g^{-1}	CD	1 A g^{-1}	0–0.5 V	6 M KOH	8
MnO_2@Ni foam/Ni–Fe-LDH	4274.4 mF cm^{-2}	CD	5 mA cm^{-2}	0–0.4 V	2 M KOH	9
$NiFe_2O_4$ nanosheet	1139 F g^{-1}	CD	5 mA cm^{-2}	0–1.2 V	6 M KOH	10
This work	2697 F g^{-1}	CD	1 A g^{-1}	0–0.55 V	1 M KOH	-
This work	1299 F g^{-1}	CV	10 mV s^{-1}	0–0.55 V	1 M KOH	-

Fig. S10. (a) CV curves and (b) specific capacitance plots of **CNF4** and the physical mixture of C_{60} and Ni–Fe-LDH at a scan rate of 10 mV s^{-1}.

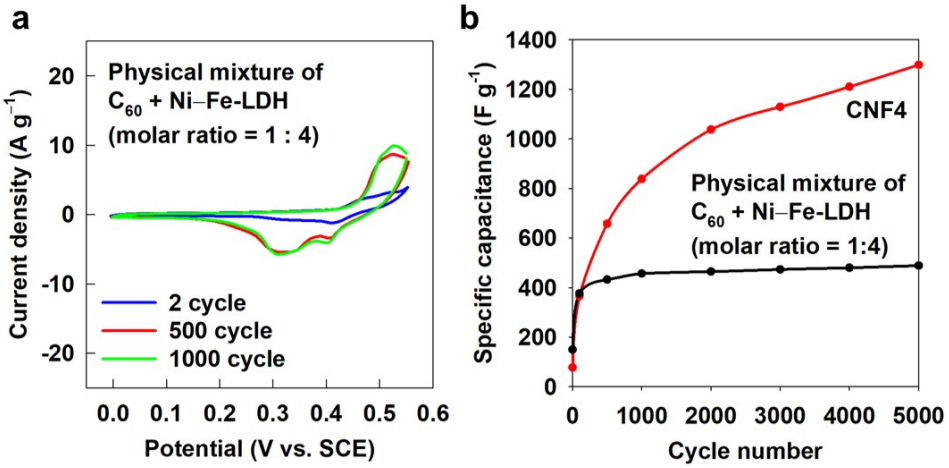


Fig. S11. Contribution ratio of capacitive- and diffusion-controlled charge storages at various scan rates for (a) Ni–Fe-LDH (b) C_{60}. Potential-dependent total and capacitive contributions of (c) Ni–Fe-LDH and (d) C_{60}.

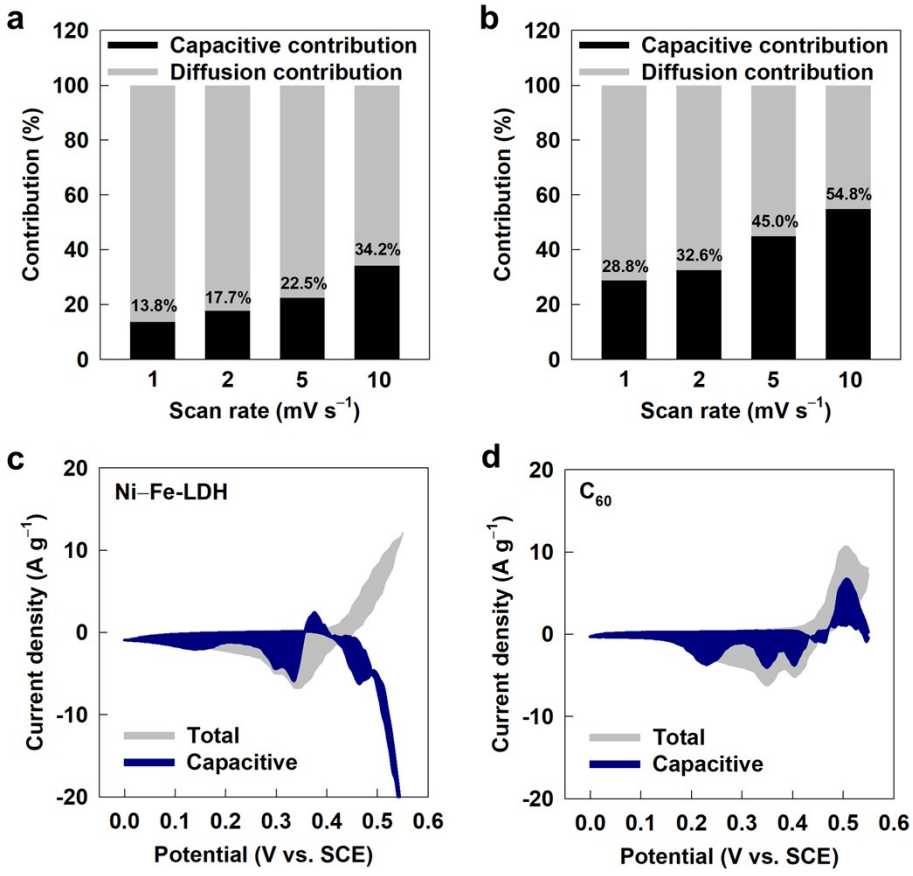


Fig. S12. Nyquist plots of the as-prepared **CNF4** nanohybrid and its 5,000-cycled derivative.

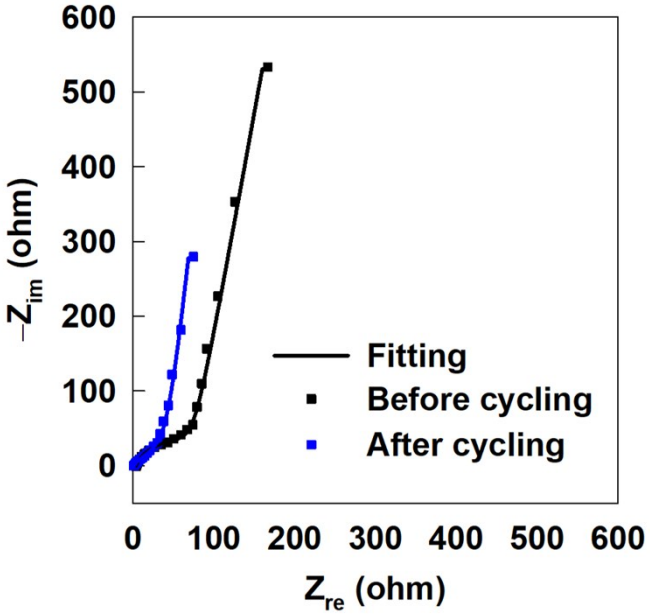
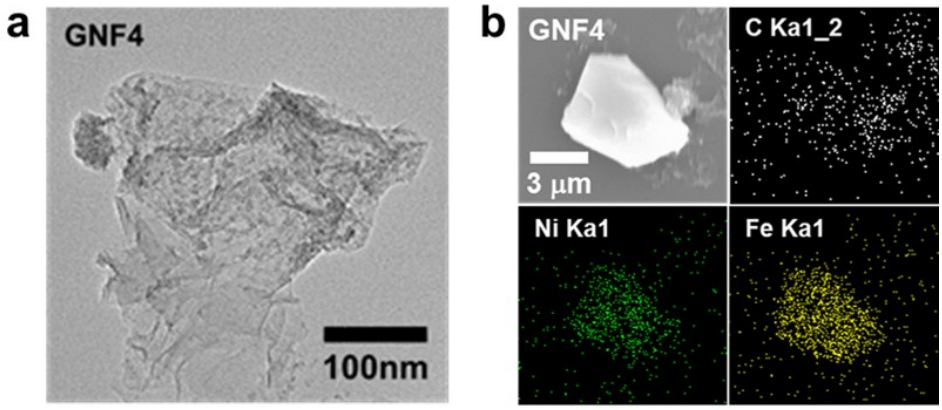


Fig. S13. (a) TEM image and (b) energy-dispersive X-ray spectroscopy (EDS)–elemental maps (right) of **GNF4** nanohybrid.



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