Assembling Ni-Fe Layered Double Hydroxide 2D Thin Films for Oxygen Evolution Electrodes

- Supporting information -

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SI-1. TEM image of Ni-Fe LDH

Figure S1 reports an additional TEM image of the Ni-Fe LDH. The low contrast of the platelets at 300 kV highlights the reduced thickness of the particles. The dark elongated particles are Ni-Fe LDH particles oriented perpendicular to the TEM grid.

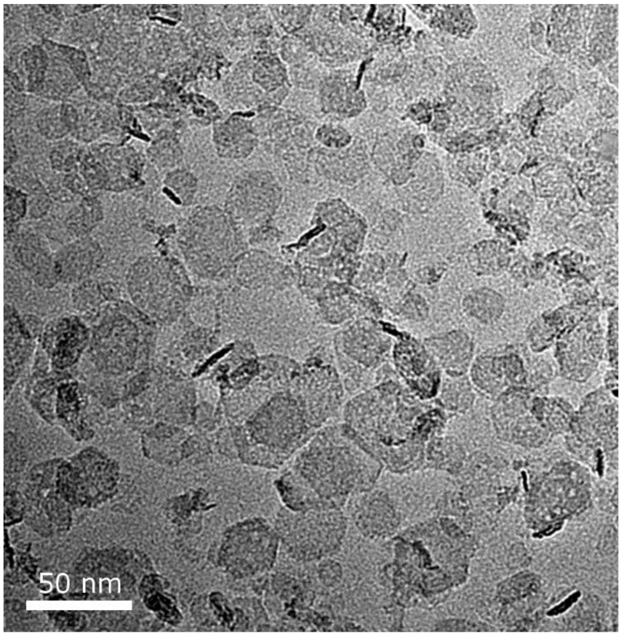

Figure S1: BF-TEM image of the NiFe LDH particles.

SI-2. Design of printed patterns

Figure S2 shows schematically the fabrication process of the electrode shown in **Fig. 4a** by using inkjet printing.

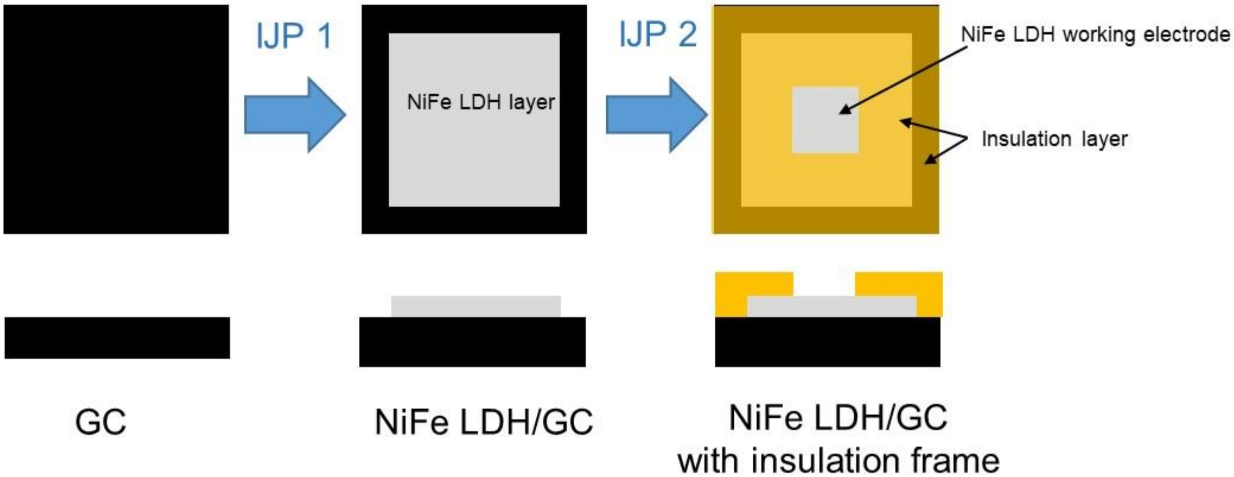

Figure S2. Schematic representation of the test electrode structure from **Fig. 4a** using multi-layer inkjet printing. Top: top view. Bottom: side view.

SI-3. Scotch tape test

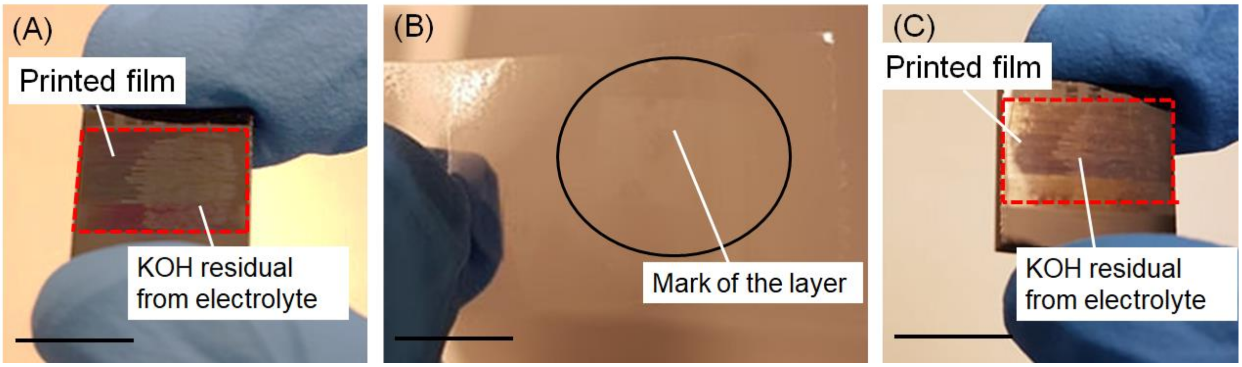

Figure S3. Result of the scotch tape test for estimating the adhesion of a Ni-Fe LDH inkjet printed film on GC. (A) Printed film before the tape test. The red dotted line indicates the printed area on GC. The image also shows the presence of a KOH residual formed after drying of the supporting electrolyte used in the electrochemical tests of this sample. (B) Photo of the tape after testing. The black circle indicates a very weak mark left by a slight detachment of material. (C) The printed film after the tape test, showing the presence of material adhered on the substrate.

SI-4. Ultra-low LDH loadings on GC

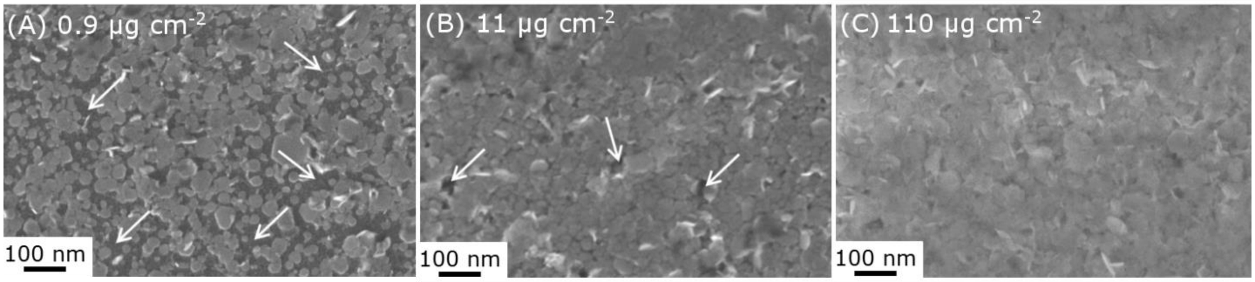

Figure S4. SEM of LDH layers with different loadings. (A) 0.9 μg cm^{-2}, (B) 11 μg·cm^{-2} and (C) 110 μg·cm^{-2}. White arrows indicate uncovered substrate spots.

SI-5. Comparison of the OER activity of the inkjet printed Ni-Fe LDH electrode with other recently reported Ni-Fe LDH materials

Table S1. List of Ni-Fe LDH from the literature with material support, synthesis method, concentration of electrolyte solution, onset potential, overpotential, Tafel slope and nominal Ni:Fe ratio.

Electrocatalyst	Support/ composite material	Synthesis	Concentration of KOH Electrolyte	Onset potential / mV vs. $E°$ (O_2,H_2O) = 1.23 V	Overpotential @ 10 mA/cm^{-2} current density / mV vs. $E°$ (O_2,H_2O) = 1.23 V	Tafel slope / mV· dec^{-1}	Nominal Ni:Fe ratio	Ref.
NiFe LDH	GC	CHFS	0.1 M	-	280	32	3:1	This work
Ni-Fe-LDH	sandblasted Ni plates, Ni foam	Batch Hydrothermal	1 M	-	280	37	optimum 7:3/3:2	1
NiFe-LDH/Fe-N-carbon nanofibers	GC	Batch Hydrothermal	1 M	-	263	81	3:1	2
NiFe-LDH	GC	Batch Hydrothermal	1 M	420 (vs MOE)	197	100	2.9:1	3
NiFe-LDH/oxygen-decorated graphene/single-walled CNT hybrids	GC	Urea assisted co-precipitation with oGSHs as the substrate	0.1 M	240	350	54	3:1	4
NiFe-LDH/CNT	GC and carbon fiber paper	Batch Solvothermal	0.1 M 1 M	270 220	290 250	35 31	5:1	5
NiFe-LDH/Ni foam	Ni foam	Potentiostatic deposition	1 M	-	269	48	1:1	6
NiFe-LDH platelets Exfoliated nanosheets	GC	Batch Hydrothermal	1 M	-	350 300	65 40	3:1 3:1	7
NiFe-LDH intercalated with SDS	Carbon fiber paper	Batch Hydrothermal	1 M	-	289	39	2:1	8
NiFe LDH/ Graphene oxide Best: rGO	Ni Foam	Batch Hydrothermal	1 M	190	205	39	3:1	9
NiFe LDH/Carbon Dot	GC	Batch Solvothermal	1 M	210	230	30	5:1	10
NiFe LDH	Ni Foam	Batch Hydrothermal	0.1 M	230	240	50	3:1	11

SI-6. Polarization curves

Polarization curves of four Ni-Fe LDH samples printed with loadings of 50 and 150 μg· cm^{-2} are shown (two curves from **Fig. 5(b)**). For clarity, only voltammograms recorded before the aging step are reported.

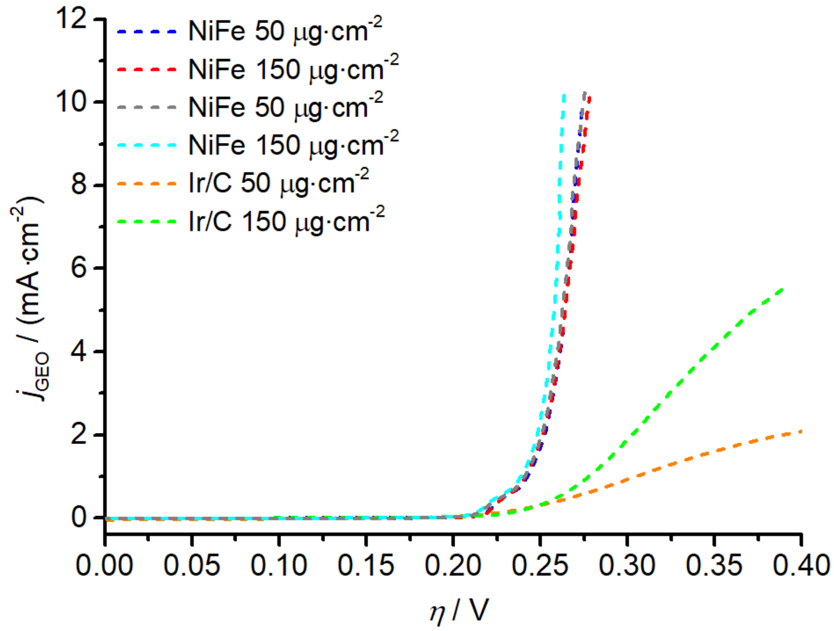

Figure S5: Additional polarization curves of samples printed with loadings of 50 (grey) and 150 (blue) μg·cm^{-2}. The remaining curves correspond to the voltammograms in **Fig. 5(b)**.

SI-7. Durability test of the inkjet printed Ni-Fe LDH/GC electrode

Chronopotentiometric experiment with the RDE were carried out for 3h with a current density of 10 mA cm^{-2} and rotating speed of 1600 rpm.

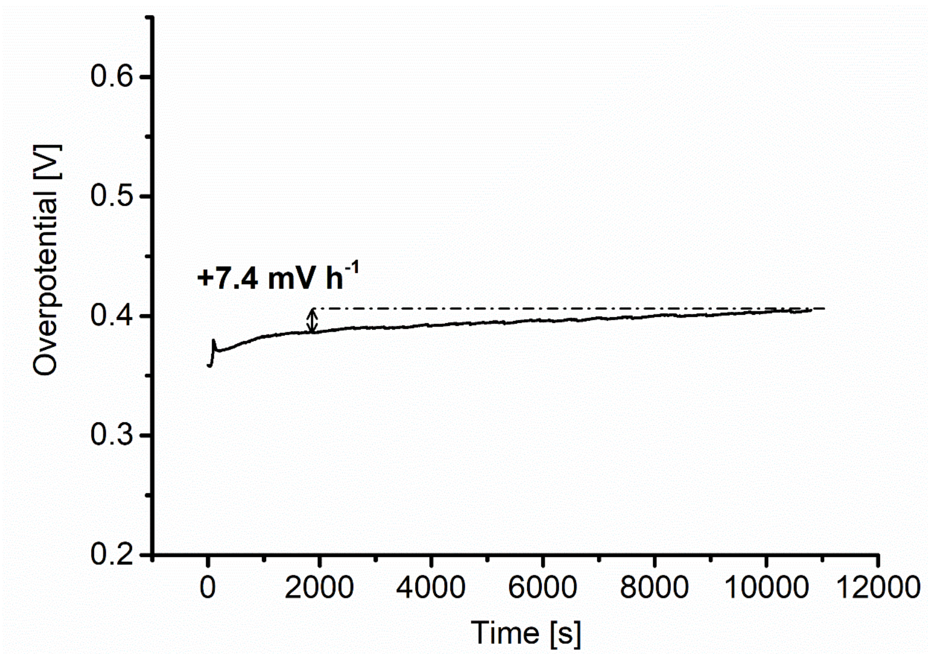

Figure S6. Chronopotentiometric durability test at 10 mA cm^{-2} performed in RDE configuration of a Ni-Fe LDH sample printed on a glassy carbon rod with a loading of 60 µg cm^{-2} previously cycled 50 times. The overpotential is not *iR*-corrected.

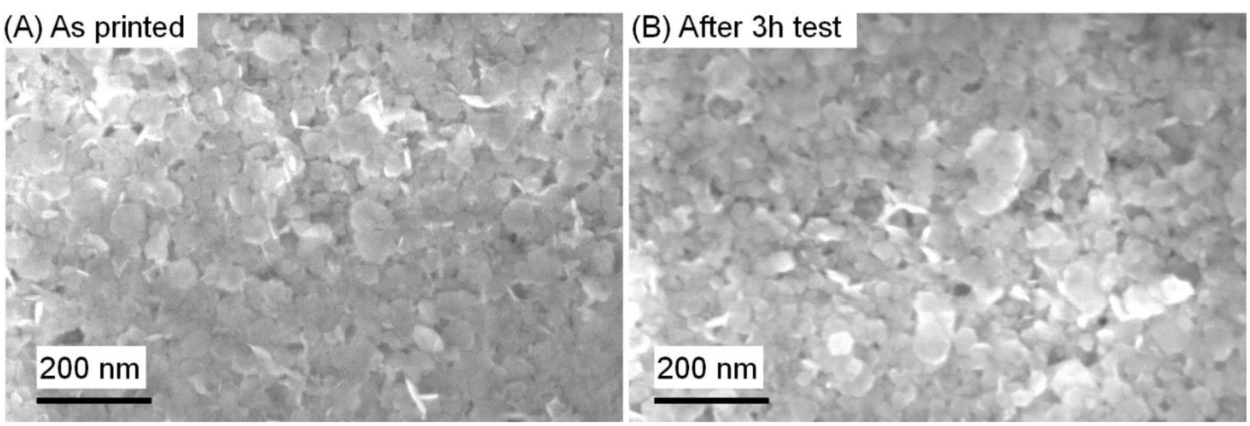

Figure S7. SEM of the Ni-Fe LDH/GC electrode before (A) and after (B) the durability test.

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