

Electronic Supplementary Information for

**Towards superior oxygen evolution through graphene barriers
between metal substrates and hydroxide catalysts**

*Hao-Fan Wang¹, Cheng Tang¹, and Qiang Zhang**

Beijing Key Laboratory of Green Chemical Reaction Engineering and Technology,
Department of Chemical Engineering, Tsinghua University, Beijing 100084, PR China

* Corresponding author.

E-mail address: zhang-qiang@mails.tsinghua.edu.cn (Q. Zhang)

¹ H.F. Wang and C. Tang contribute equally

I. Supplementary figures

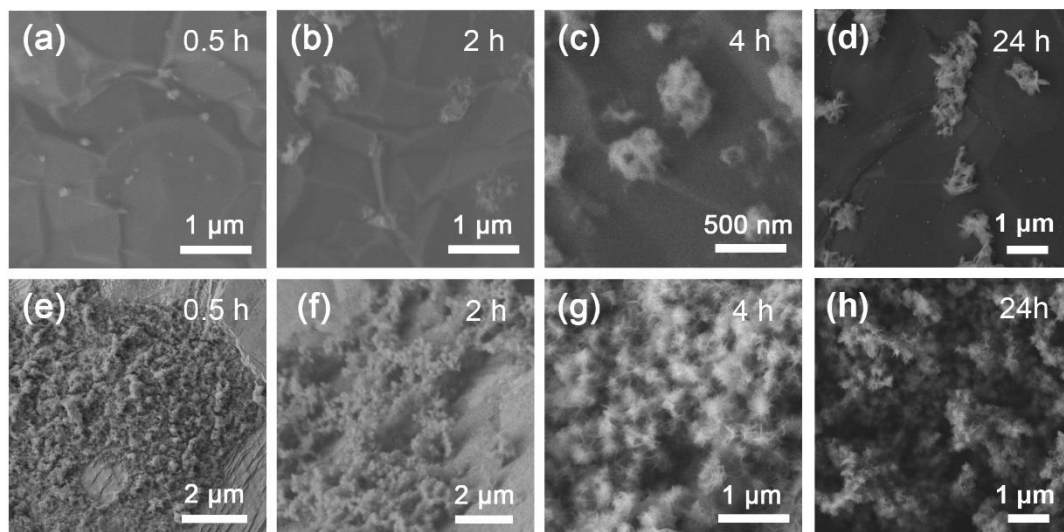


Fig. S1. (a-d) SEM images of LDH/G/Ni with the LDH growth duration of 0.5, 2.0, 4.0, and 24.0 h. (e-h) SEM images of LDH/Ni with the LDH growth duration of 0.5, 2.0, 4.0, and 24.0 h.

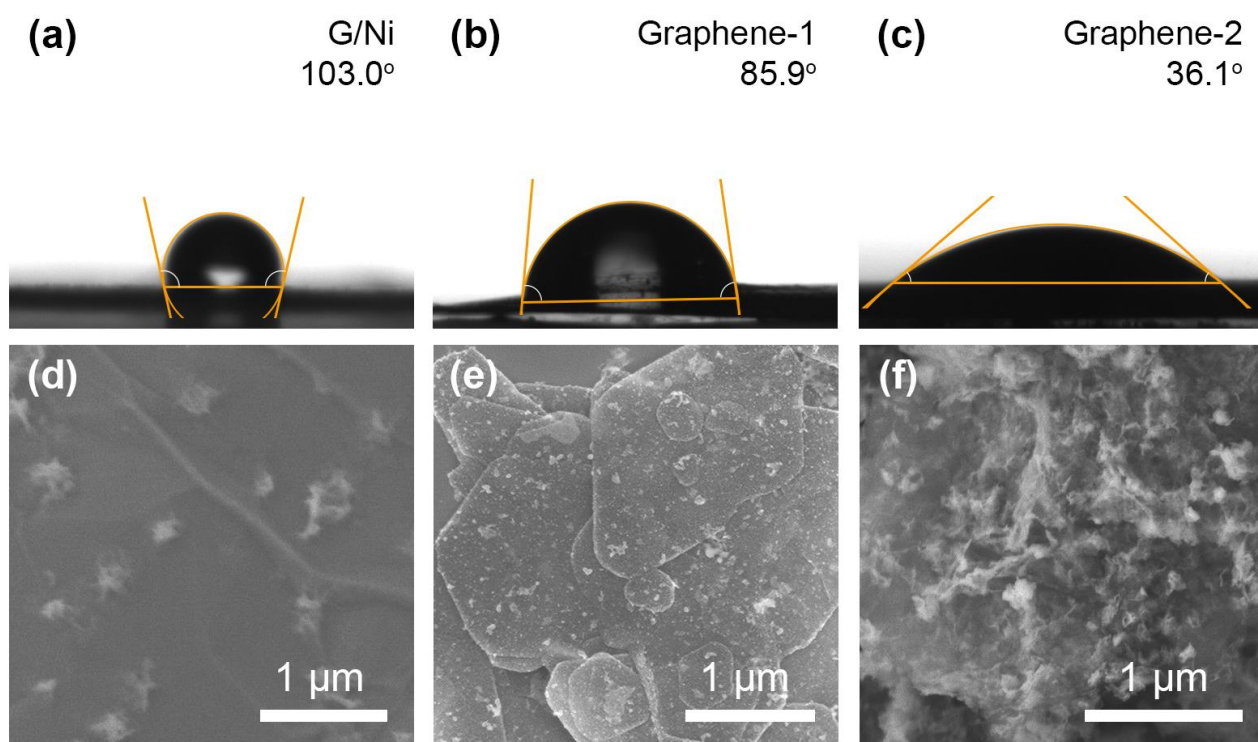


Fig. S2. (a-c) The contact angles of LDH/graphene composites with LDHs grown on G/Ni, Graphene-1, and Graphene-2. (d-f) The morphology of LDHs grown on G/Ni, Graphene-1, and Graphene-2. Graphene-1 was synthesized by a MgO-templated CVD method at 950 °C for 10 min using methane as carbon source. After purification, the graphene with a contact angle of 85.9 ° was obtained by further mixing with concentrated sulfuric acid at 10 °C for 10 h. Graphene-2 (thermal reduced graphene oxide (GO)) with a contact angle of 36.1 ° was prepared with graphite as raw material. Graphite was oxidized to GO by a modified Hummer's method, then the GO was kept at 250 °C for 20 min in high vacuum (below 5 Pa) to obtain Graphene-2. The LDHs get dense and continuous with decrease of the contact angle, indicating that the hydrophobicity of graphene increases the LDH nucleation resistance and facilitates the loose distribution of LDHs.

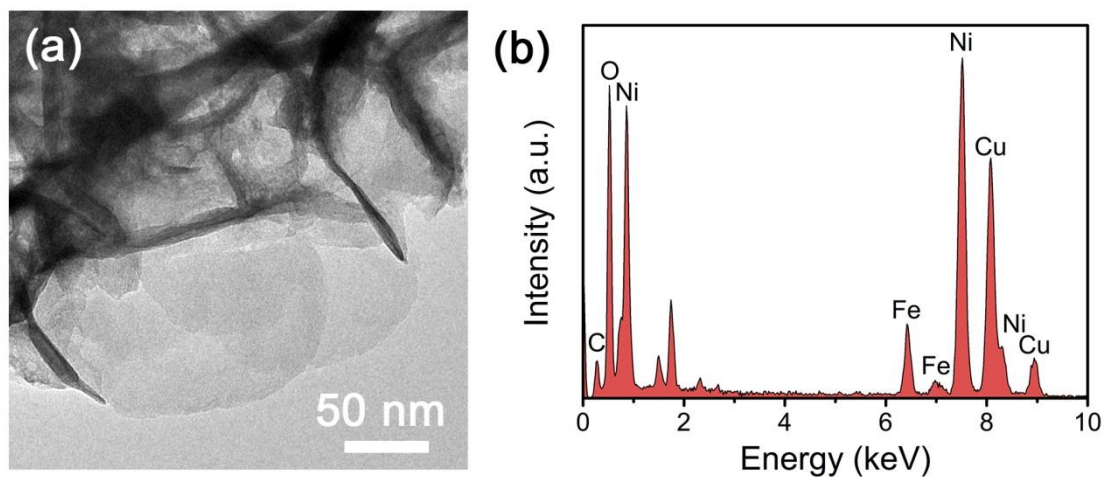


Fig. S3. (a) TEM image of p-NiFe LDH. (b) EDS pattern of p-NiFe LDH in the region presented in Fig. S3a. The nano-structure of p-NiFe LDHs resembles the LDH sheets on LDH/G/Ni. The Ni:Fe mole ratio of p-NiFe LDH is 5.9:1, also approximate to that of LDHs in LDH/G/Ni.

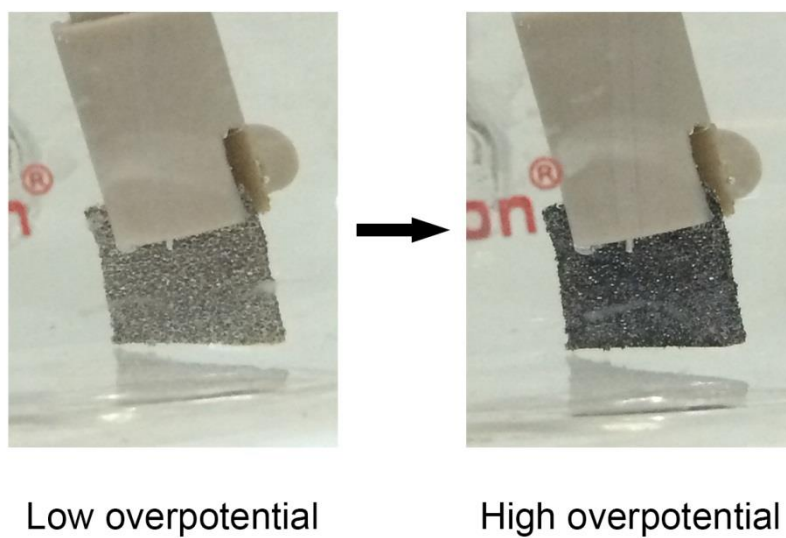


Fig. S4. The morphology evolution of G/Ni during the OER process, indicating the oxidation of Ni at a high overpotential of 0.5 V.

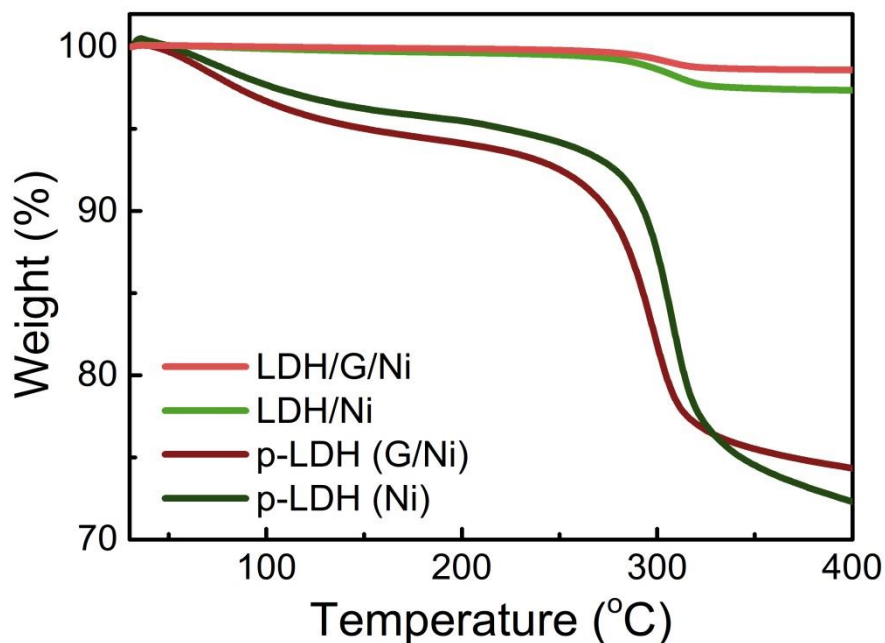


Fig. S5. TGA plots of LDH/G/Ni, LDH/Ni, and the respective precipitate LDHs. The p-LDH (G/Ni) stands for the LDH precipitate generated in the synthesis process of LDH/G/Ni, and p-LDH (Ni) was named in the same way. The LDHs grown on substrate and the LDH precipitate are regarded as the same in composition. Assuming that the mass of LDH/G/Ni was m , and the weight loss were $a\%$ and $b\%$ for LDH/G/Ni and p-LDH (G/Ni), then the loading amount of LDH on LDH/G/Ni was $m \cdot a/b$. The computing method is the same for LDH/Ni. The loading amount of LDHs on LDH/G/Ni and LDH/Ni is 2.18 and 3.63 mg cm^{-2} , respectively.

LDH/Ni
20.3°

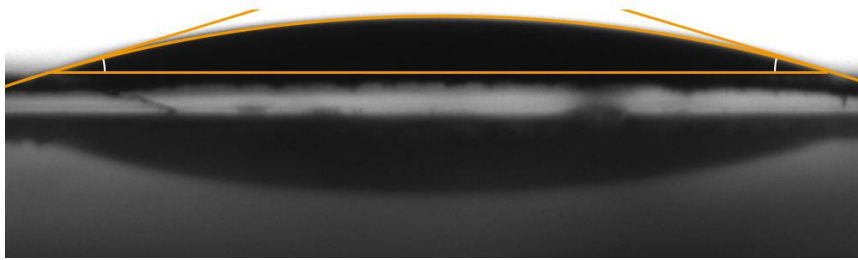


Fig. S6. The contact angle of LDH/Ni sample.

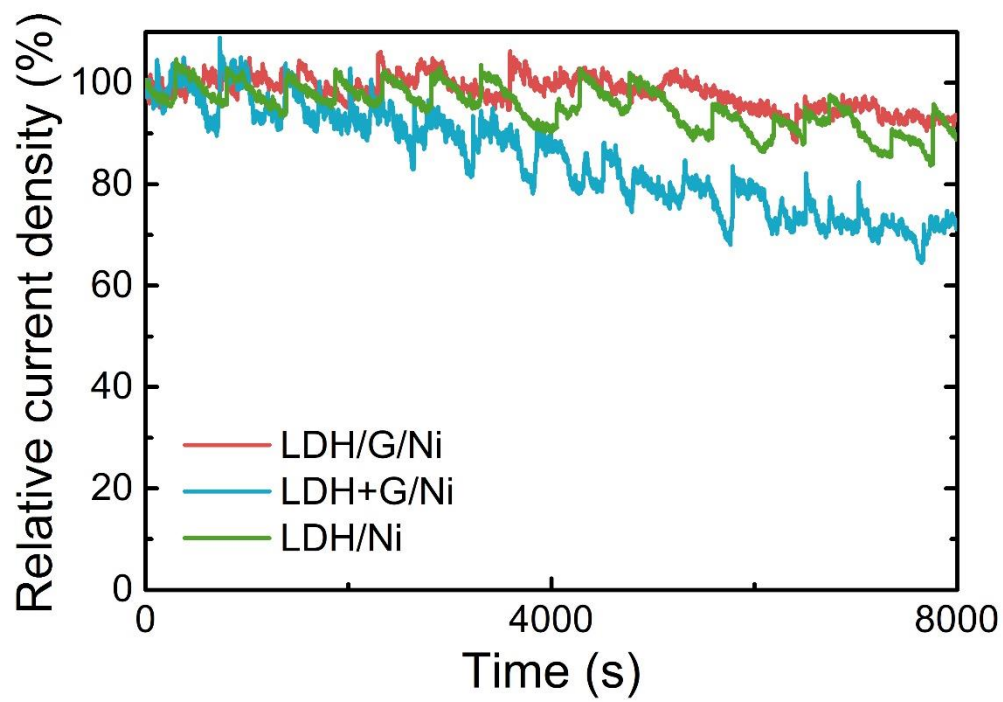


Fig. S7. Chronoamperometric response of LDG/G/Ni, LDH+G/Ni and LDH/Ni at initial current density of 10.0 mA cm⁻² in 0.10 M KOH.

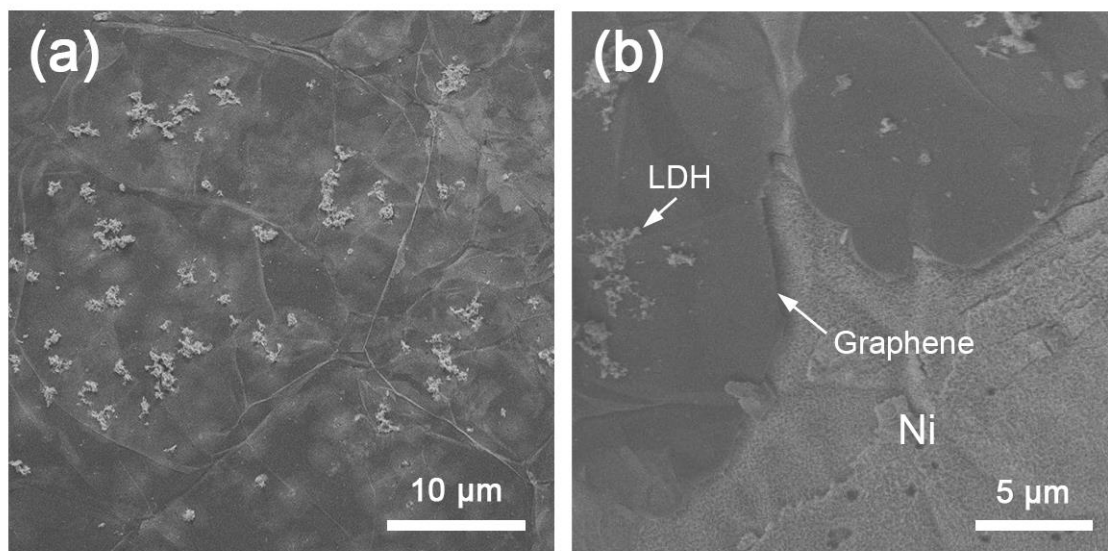


Fig. S8. The SEM images of LDH/G/Ni after long period of OER catalysis. (a) The morphology remains the same as the initial LDH/G/Ni in most regions. (b) Graphene layer peeled off and bare Ni was exposed at few edge areas.

II. Supplementary table

Table S1. A summary of the OER performances of on transition metal compounds based catalysts^{a)}

Samples	η_{onset} (mV)	η_{10} (mV)	j_{350} (mA cm ⁻²)	Tafel slope (mV dec ⁻¹)	Ref.
LDH/G/Ni	240	325	34.8	44	This work
LDH/Ni	250	350	10.3	51	
G/Ni	300	399	2.6	52	
Ni-Fe LDH/SWCNT	270	316	-	35	[1]
NG/NiCo ₂ O ₄	323	457	3.5	156	[2]
3D NF/PC/AN	297	510	-	-	[3]
MWCNT/Ni(OH) ₂	350	474	0	87	[4]
3D NiFe LDH	230	250	-	50	[5]
CQD/NiFe LDH	260	305	-	35	[6]
G-Co ₃ O ₄	206	359	9	67	[7]
Co ₃ O ₄ C-NA	240	290	28	70	[8]
Ni@NiCoOH	310	460	0.5	65	[9]
α -Ni(OH) ₂	310	331	0.26	42	[10]
NiCo-LDH	290	410	3.5	113	[11]
Zn-Co-LDH-100	330	510	0	83	[12]

a) All these OER performances were tested in 0.1 M KOH solution.

“-” represents that the value can't be obtained from the literature.

III. Supplementary references

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