

## Electronic Supplementary Information

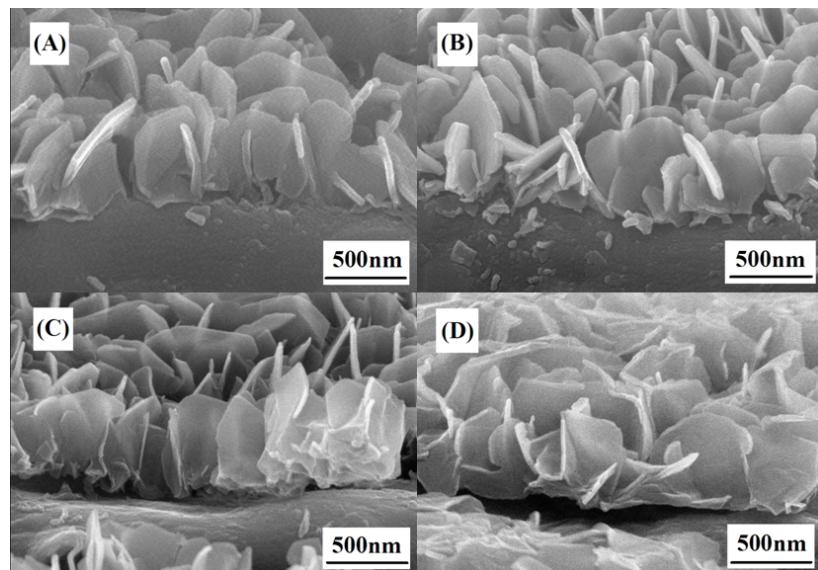
### Engineering ZnCo-Layered Double Hydroxide Nanowalls toward High-Efficiency Electrochemical Water Oxidation

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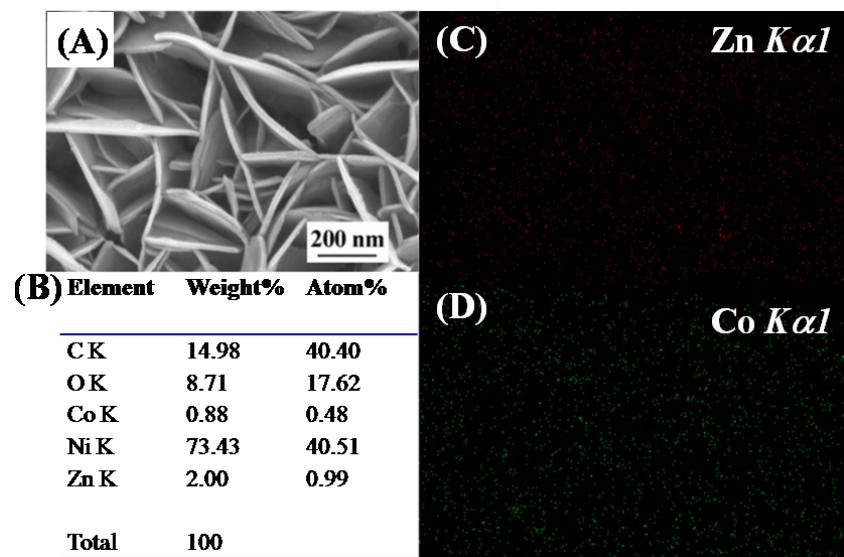
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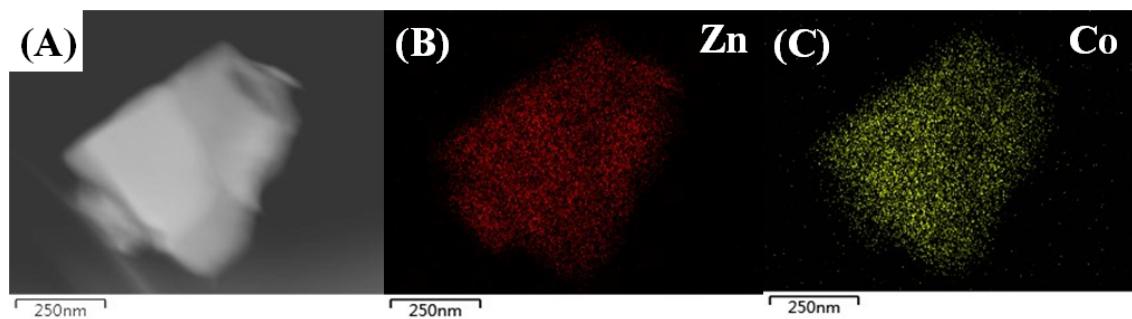
**Fig. S1** Digital photo of ZnCo-LDH-100 film directly grown on a flexible Ni foil



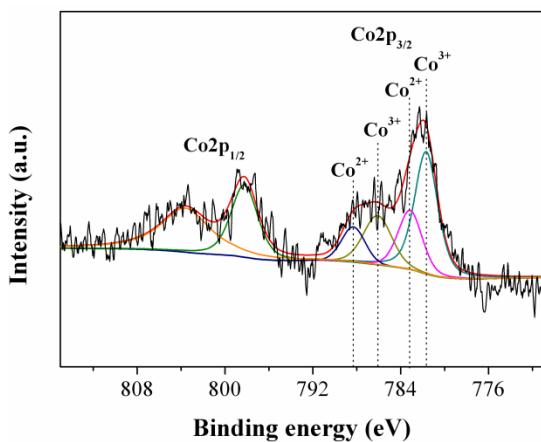
**Fig. S2** Cross-sectional SEM images of ZnCo-LDH nanowalls (A) ZnCo-LDH-50, (B) ZnCo-LDH-100, (C) ZnCo-LDH-150, (D) ZnCo-LDH-180. The film thickness is approximately 480, 510, 570 and 600 nm, respectively.



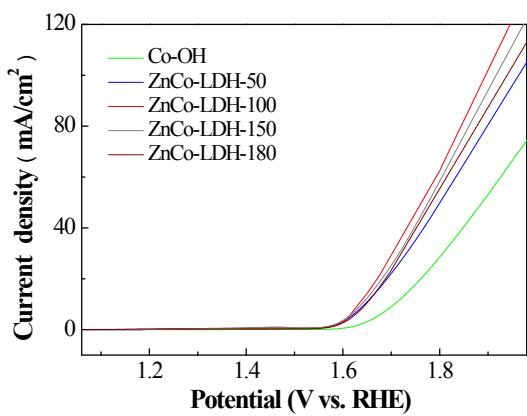
**Fig. S3** (A) SEM image of ZnCo-LDH-100 sample, (B) the elemental analyses, (C) and (D) EDX mapping of Zn and Co elements



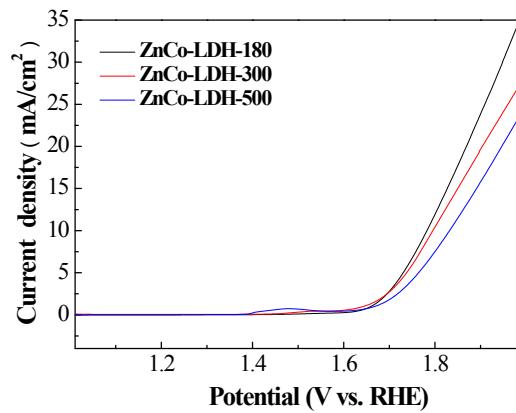
**Fig. S4** HAADF image (A) and EDX mappings (B) and (C) of ZnCo-LDH-100



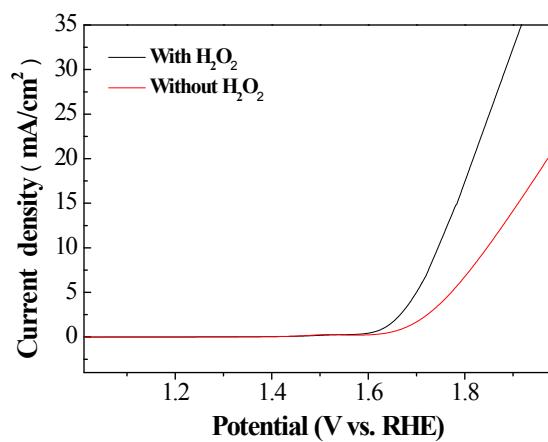
**Fig. S5** XPS core level spectra of Co<sup>2p</sup> in the ZnCo-LDH-100



**Fig. S6** LSV curves tested in 1 M KOH solution with ZnCo-LDH films and Co-OH as electrocatalysts.



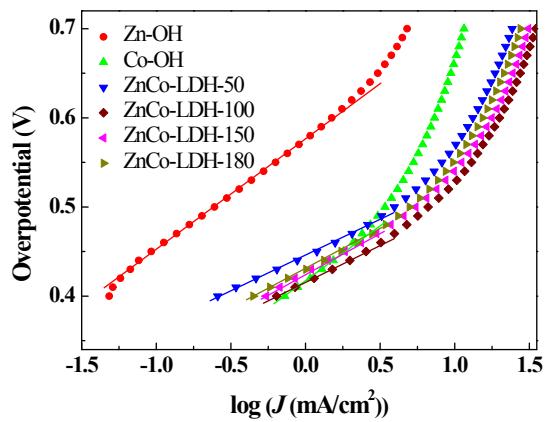
**Fig. S7** LSV curves tested in 0.1 M KOH solution with ZnCo-LDH films at longer deposition time



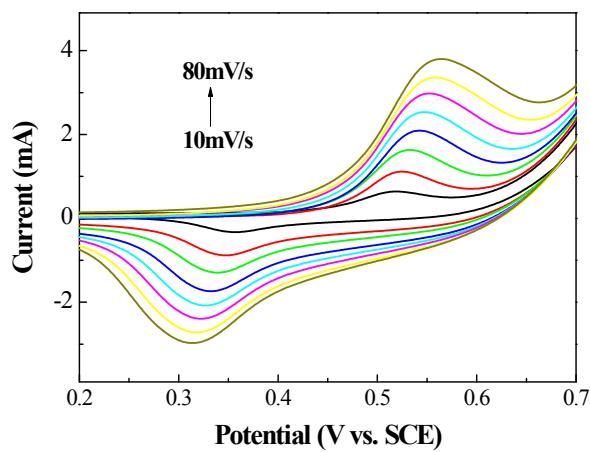
**Fig. S8** LSV curves of ZnCo-LDH-100 films with addition of  $\text{H}_2\text{O}_2$  (black) and without  $\text{H}_2\text{O}_2$  (red) during the preparation of films, the LSV was tested in 0.1 M KOH solution.



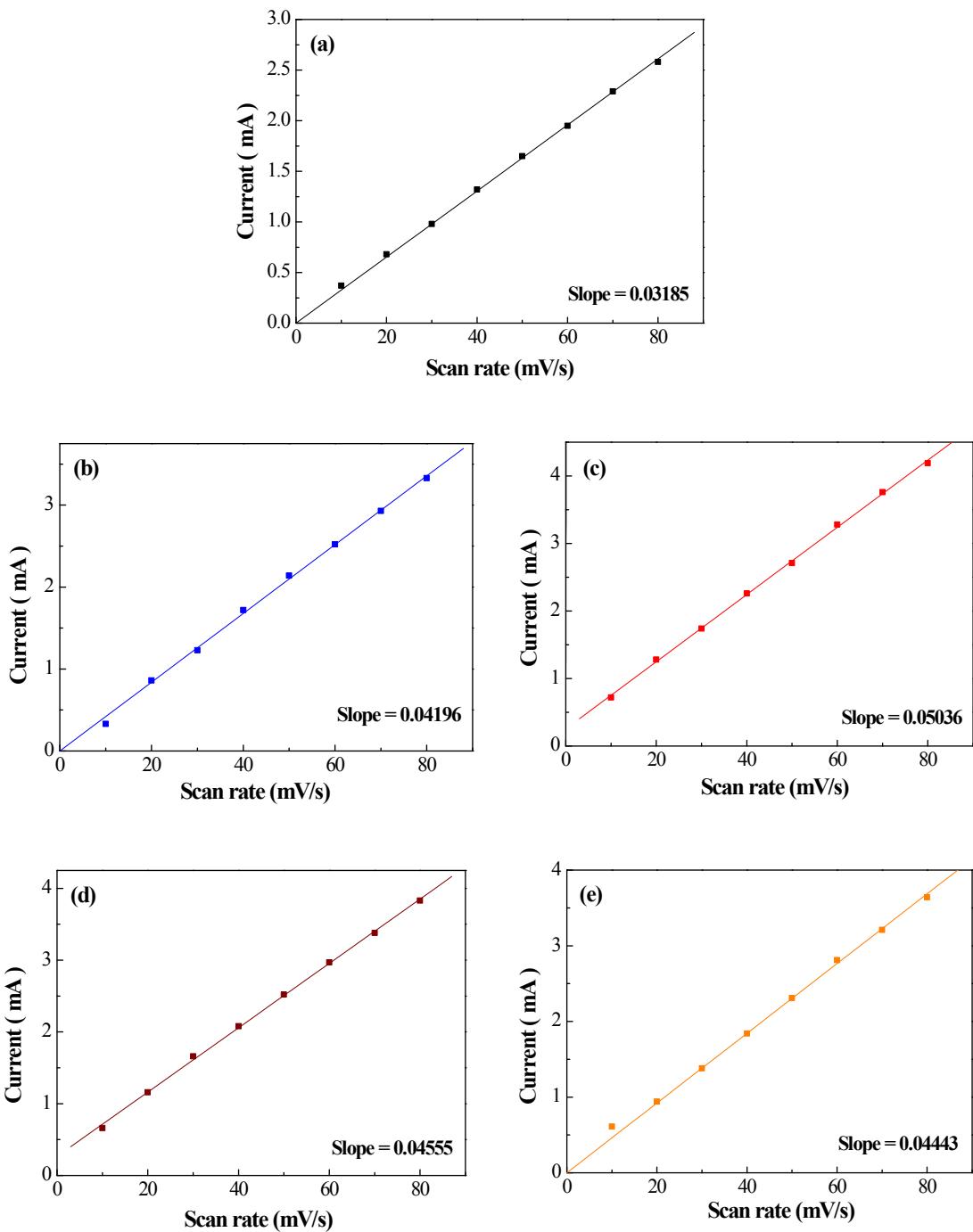
**Fig. S9** Digital photos of ZnCo-LDH-100 films on Ni substrate before (left) and after (right) OER operation of 2 hr at an applied potential of 1.667 V



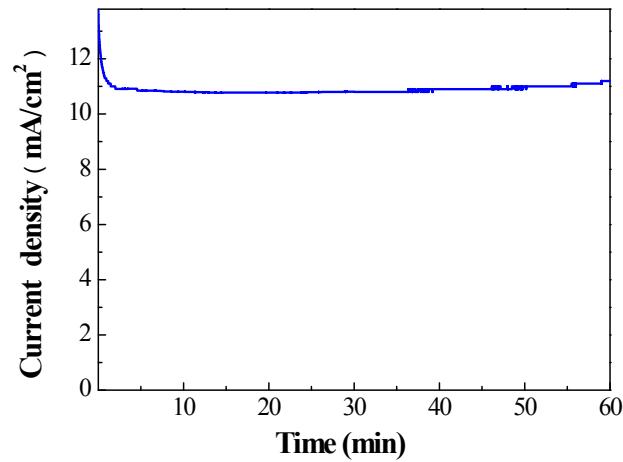
**Fig. S10** Tafel plots of ZnCo-LDH electrocatalysts and the control samples



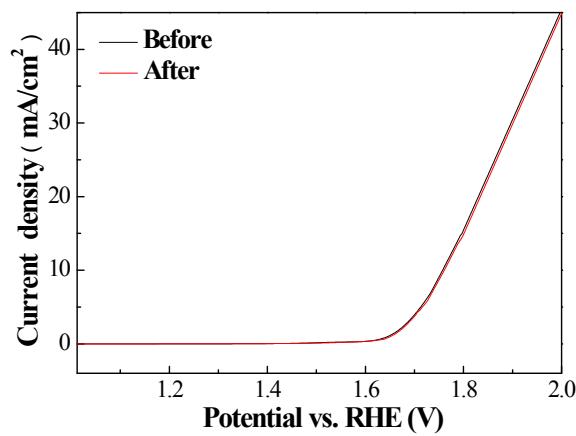
**Fig. S11** Cyclic voltammogram curves of ZnCo-LDH-150 under different scan rate tested in 0.1 M KOH solution.



**Fig. S12** Linear relationship of the peak current of  $\text{Co}^{3+}/\text{Co}^{2+}$  oxidation wave with the scan rate (a) Co-OH, (b) ZnCo-LDH-50, (c) ZnCo-LDH-100, (d) ZnCo-LDH-150, (e) ZnCo-LDH-180



**Fig. S13** Current-time curve during OER with ZnCo-LDH-100 as electrocatalyst at an applied potential of 1.767 V



**Fig. S14** LSV curves of ZnCo-LDH-100, before OER (black) and after 1 h OER operation (red) (tested in 0.1 M KOH)

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**Table S1** Comparison of ZnCo-LDH electrocatalysts based on a mass activity

Catalyst	ZnCo-LDH-50	ZnCo-LDH-100	ZnCo-LDH-150	ZnCo-LDH-180
Mass (mg) <sup>a</sup>	0.22	0.32	0.37	0.42
Mass activity @ $\eta = 400\text{mV}$ (A/g) <sup>b</sup>	2.3	5.8	3.2	2.1
Mass activity @ $\eta = 450\text{mV}$ (A/g)	11	21	11	8.3
Mass activity @ $\eta = 500\text{mV}$ (A/g)	36	51	32	24

<sup>a</sup> The mass was calculated by weighing the mass of Ni foil before and after the electrodeposition reactions.

<sup>b</sup> The current values were obtained from LSV curves tested in 0.1 M KOH solution.

**Table S2** Electrochemical performance comparison of ZnCo-LDH and Co<sub>3</sub>O<sub>4</sub> OER catalysts reported in the literatures

Catalyst	Current density(mA/cm <sup>2</sup> )	η (mV)	pH	TOF(s <sup>-1</sup> )	Refs
ZnCo-LDH-100	0.67	300	14	0.0245	This work
ZnCo-LDH-100	0.917	330	14	0.045	This work
Co <sub>3</sub> O <sub>4</sub>	/	328	14	0.0187	[1]
Co <sub>3</sub> O <sub>4</sub>	/	300	14	0.04	[2]

**Table S3** Electrochemical performance comparison of ZnCo-LDH and IrO<sub>2</sub> or RuO<sub>2</sub> OER catalysts reported in the literatures

Catalyst	Current density (mA/cm <sup>2</sup> )	η (mV)	Tafel slope (mV/dec)	TOF(s <sup>-1</sup> )	pH	Refs
ZnCo-LDH-100	0.26	330	83	0.025	13	This work
ZnCo-LDH-100	0.67	300	61	0.0245	14	This work
IrO <sub>2</sub>	0.5	300	/	/	13	[3]
RuO <sub>2</sub>	0.7	300	/	/	13	[3]
IrO <sub>2</sub>	11 <sup>a</sup>	300	/	0.02	13	[3]
IrO <sub>x</sub>	15 <sup>a</sup>	300	49	0.009	14	[4]

<sup>a</sup> The values are in unit of A/g.

## References

1. A. J. Esswein, M. J. McMurdo, P. N. Ross, A. T. Bell and T. Don Tilley, *J. Phys. Chem. C*, 2009, **113**, 15068-15072.
2. N. H. Chou, P. N. Ross, A. T. Bell and T. D. Tilley, *ChemSusChem*, 2011, **4**, 1566-1569.
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4. L. Trotochaud, J. K. Ranney, K. N. Williams and S. W. Boettcher, *J. Am. Chem. Soc.*, 2012, **134**, 17253-17261.