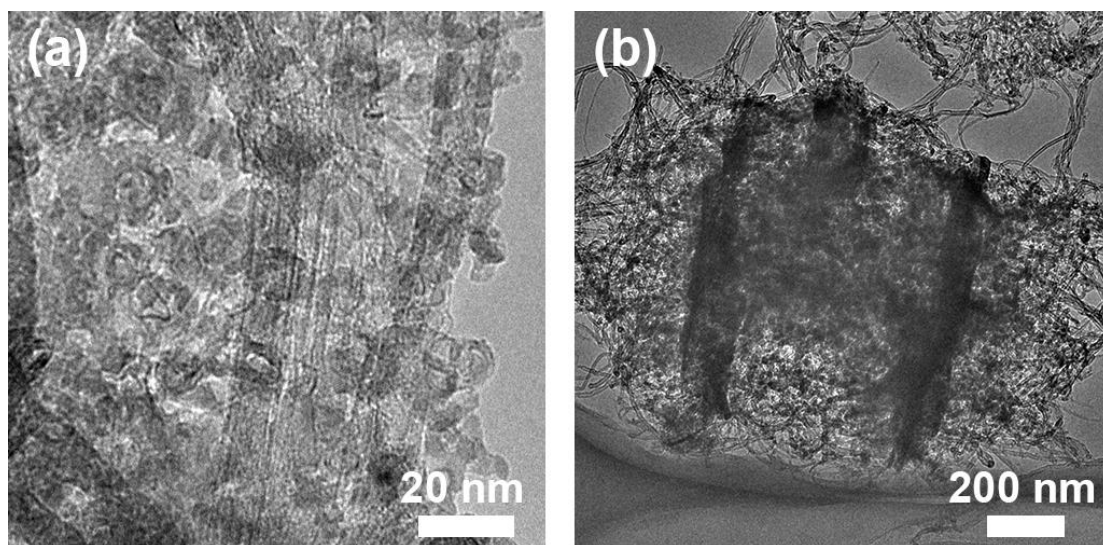
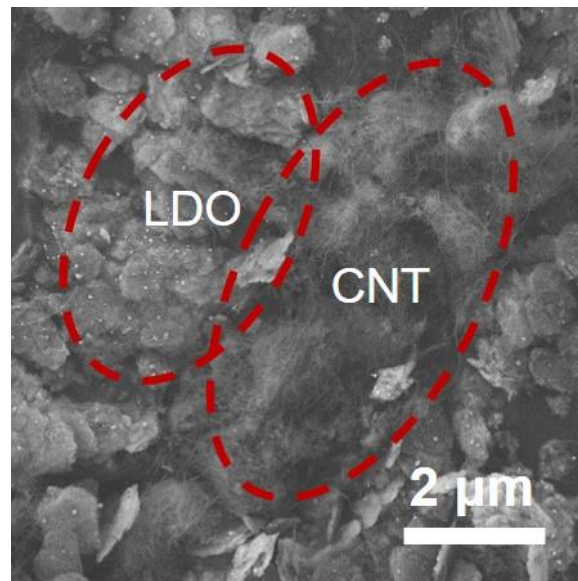


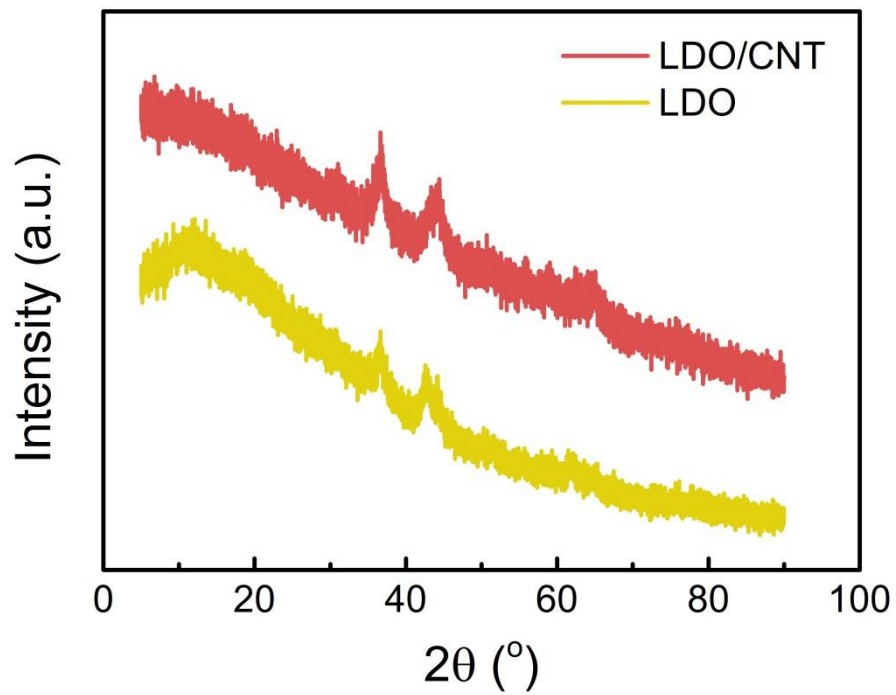
**Fig. S1.** SEM image of CoFeMgAl LDH flakes.



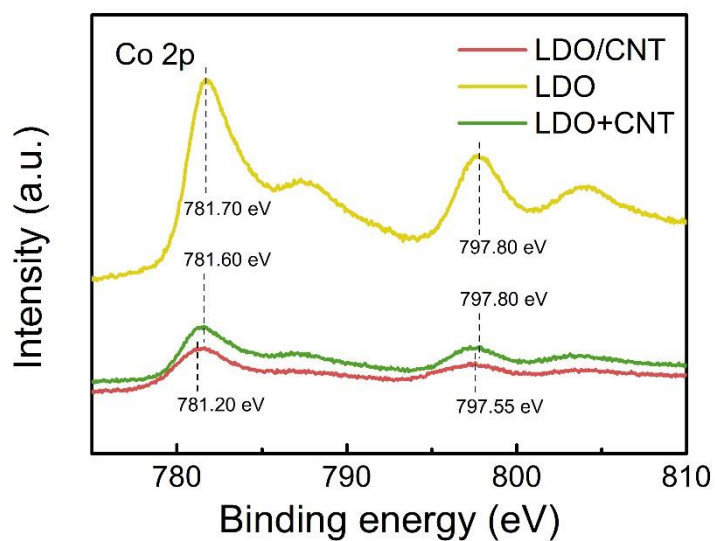
**Fig. S2.** (a) High resolution, (b) low resolution TEM images of LDO/CNT hybrids .



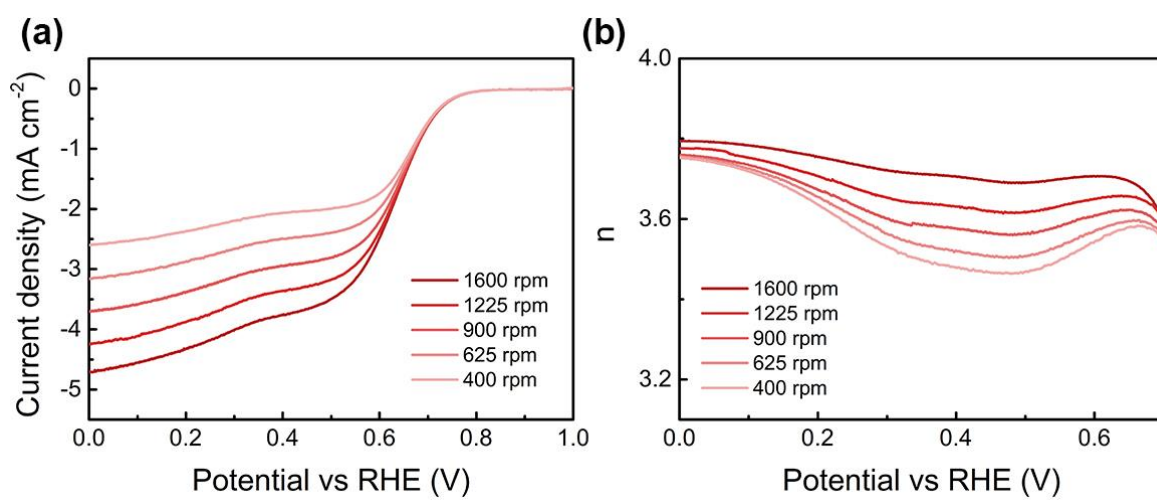
**Fig. S3.** SEM image of LDO+CNT mixture.



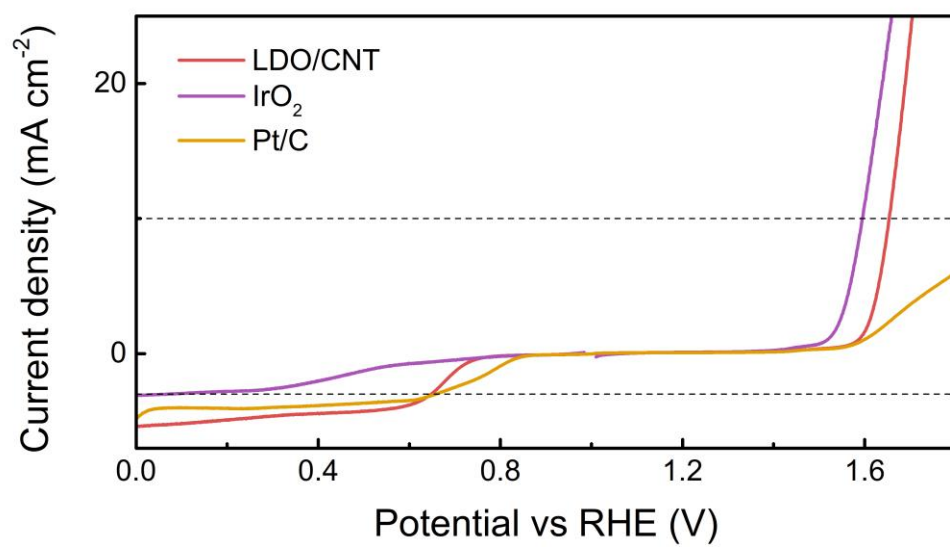
**Fig. S4.** XRD patterns of LDO/CNT hybrids and LDO flakes.



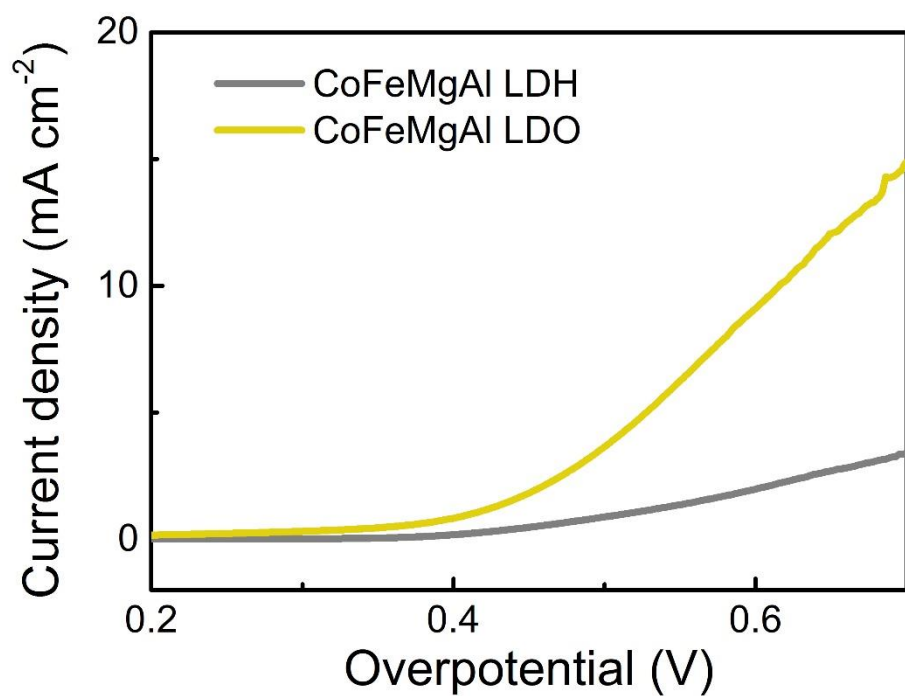
**Fig. S5.** The Co 2p spectra of LDO/CNT, LDO and LDO+CNT. Compared with LDO, LDO/CNT exhibited a shift to lower binding energy, while the peak position of LDO+CNT didn't exhibit obvious shift, indicating the chemical bond interaction between LDO and CNT in LDO/CNT, and the line-face configuration of LDO+CNT leads to an insufficient contact. The weaker peak intensity of LDO/CNT than LDO+CNT was probably due to the carbon coating on the Co-contained active centres in LDO/CNT.



**Fig. S6.** ORR (a) LSV plots and (b) electron transfer number of LDO/CNT catalyst at different rotating rates.

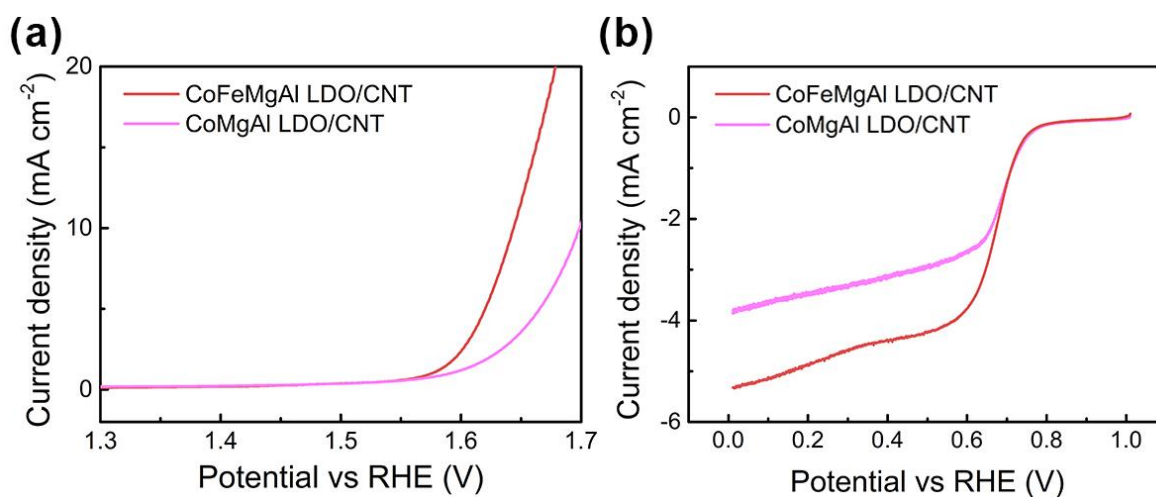


**Fig. S7.** The overall LSV plots of LDO/CNT hybrid, IrO<sub>2</sub>, and Pt/C electrocatalysts at the scan rate of 10.0 mV s<sup>-1</sup> in O<sub>2</sub>-saturated 0.10 M KOH solution.

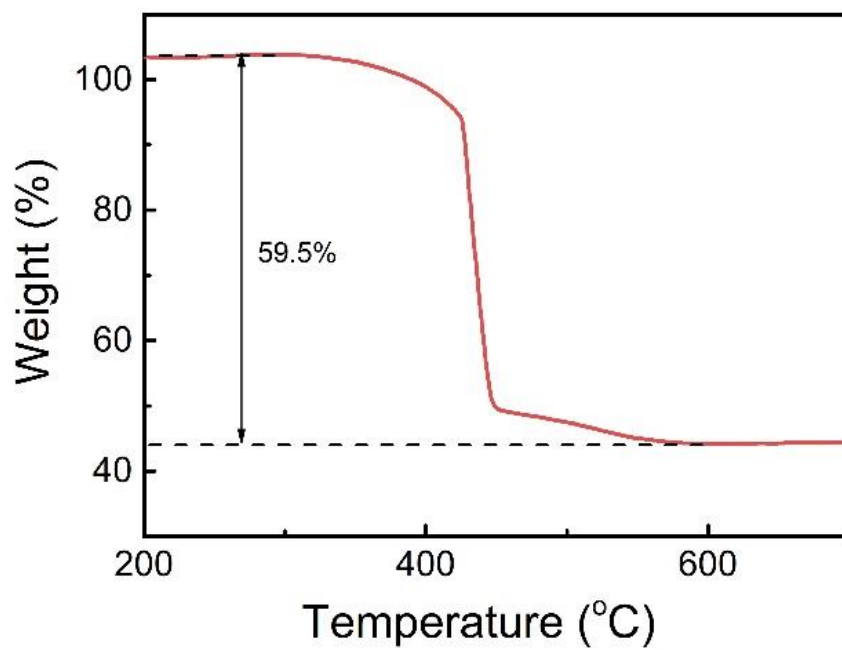


**Fig. S8.** OER LSV plots of LDH and LDO at the scan rate of 10.0 mV s<sup>-1</sup> in O<sub>2</sub>-saturated 0.10 M KOH solution.





**Fig. S9.** (a) OER and (b) ORR LSV plots of CoFeMgAl LDO/CNT hybrids and CoMgAl LDO/CNT hybrids at the scan rate of 10.0 mV s<sup>-1</sup> in O<sub>2</sub>-saturated 0.10 M KOH solution. The Co content in the CoMgAl LDO was the same as the Co and Fe total content in CoFeMgAl LDO.



**Fig. S10.** TGA plots of LDO/CNT hybrids. The weight loss at 400-600 °C indicates that the mass fraction of CNT in the original LDO/CNT catalyst is 59.5%.

**Table S1.** Summary of  $E_{10}$ ,  $E_3$ , and  $\Delta E$  of all electrocatalysts in this contribution.

<b>Catalysts</b>	$E_{10}$ (V)	$E_3$ (V)	<b>Potential gap <math>\Delta E</math> (V)</b>
LDO/CNT	1.64	0.65	0.99
LDO	1.77	0.06	1.71
CNT	1.81	0.43	1.38
LDO+CNT	1.73	0.39	1.34
IrO <sub>2</sub>	1.60	0.07	1.53
Pt/C	1.94	0.66	1.28