Supporting Information

Construction of rare-earth-modified layered double hydroxide heterostructures and enhancement of overall water splitting efficiency

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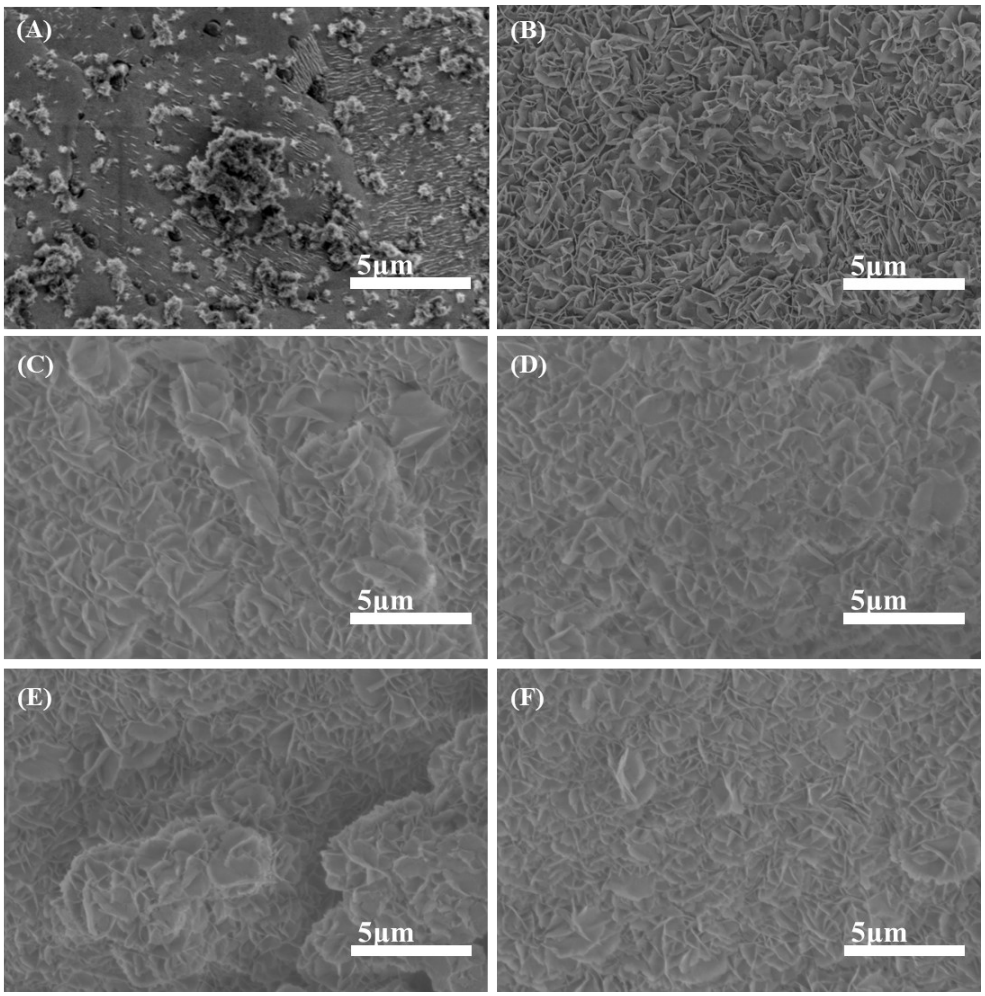

Figure S1. SEM images of (A) TiO_2@C, (B) CoCe-LDH, (C) $Co_{0.1}Ce_{0.1}$-LDH-TiO_2@C, (D) $Co_{0.1}Ce_{0.05}$-LDH-TiO_2@C, (E) $Co_{0.1}Ce_{0.005}$-LDH-TiO_2@C, (F) $Co_{0.1}Ce_{0.001}$-LDH-TiO_2@C.

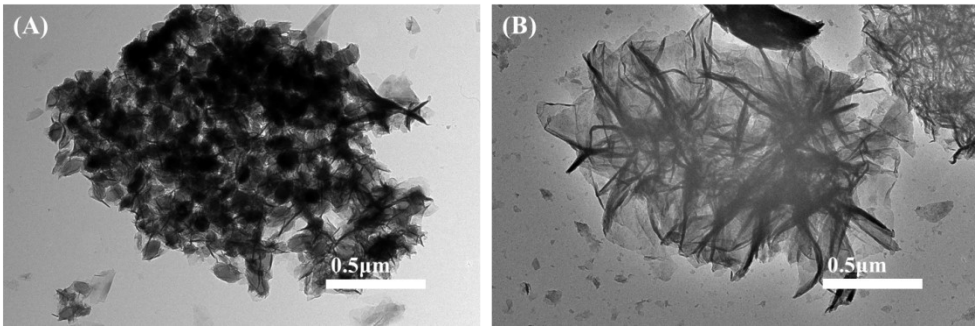

Figure S2. TEM images of (A) TiO_2@C, (B) CoCe-LDH.

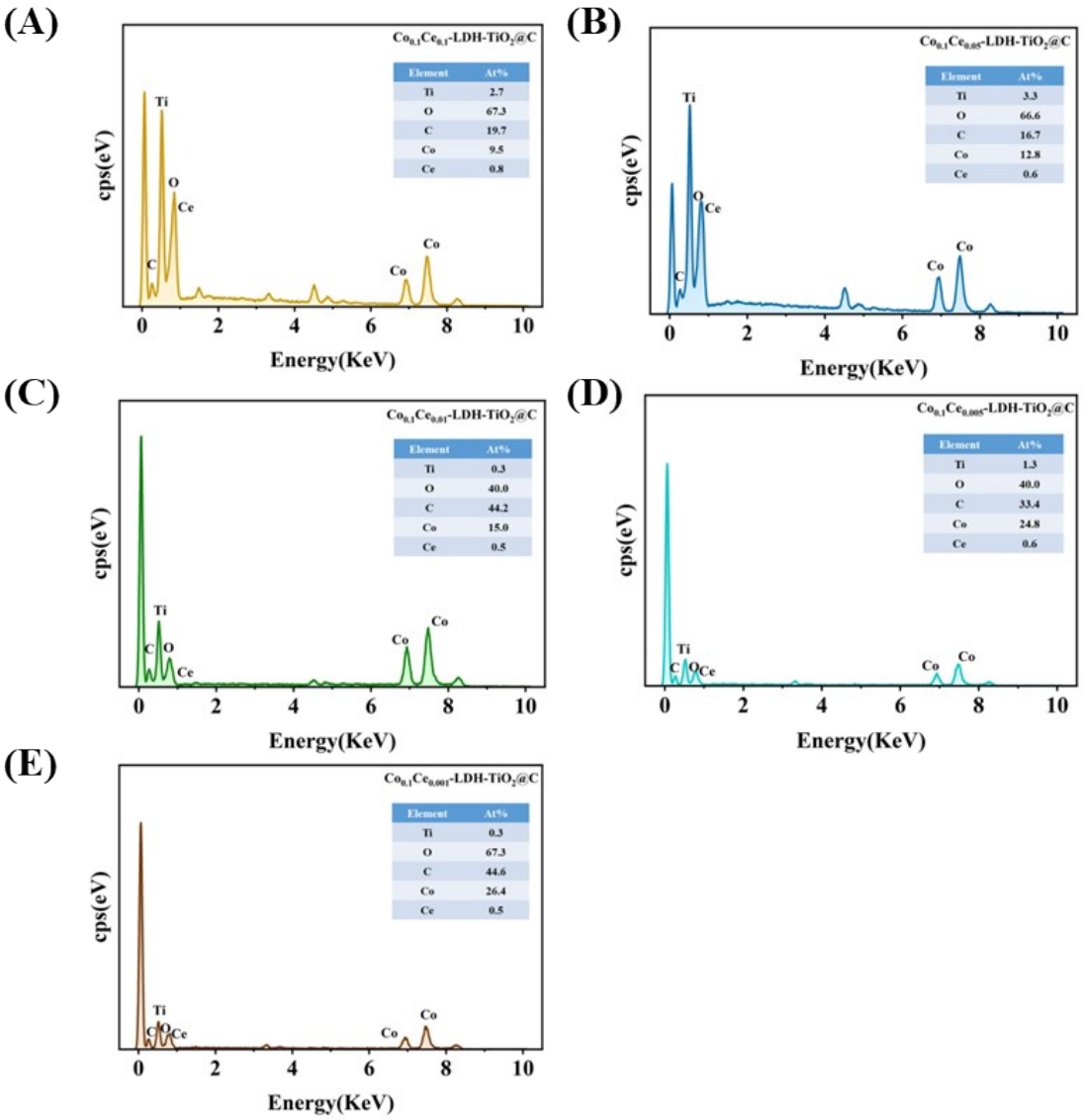

Figure S3. The EDS spectrum of aa (A) $Co_{0.1}Ce_{0.1}$-LDH-TiO_2@C, (B) $Co_{0.1}Ce_{0.05}$-LDH-TiO_2@C, (C) $Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C, (D) $Co_{0.1}Ce_{0.005}$-LDH-TiO_2@C, (E) $Co_{0.1}Ce_{0.001}$-LDH-TiO_2@C.

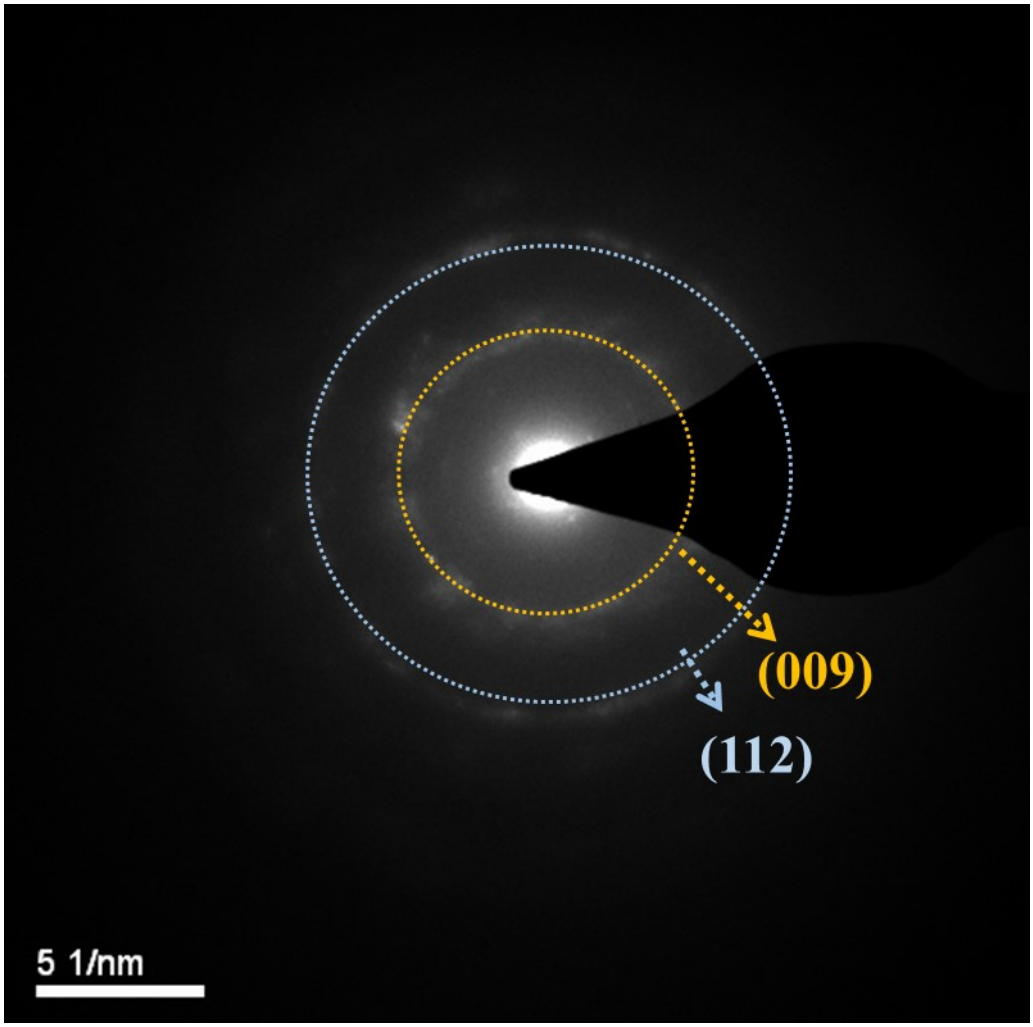

Figure S4. SAED image of $Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C.

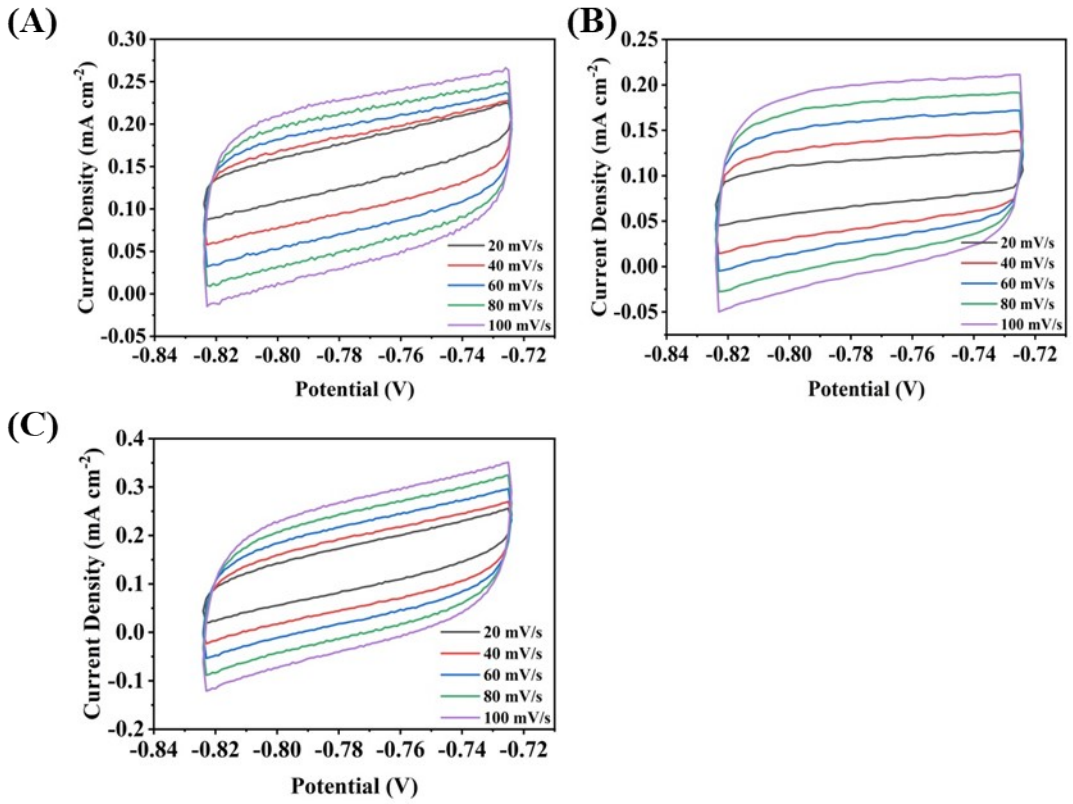

Figure S5. CV of (A) TiO_2@C, (B) CoCe-LDH and (C) $Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C at various scan rates for HER.

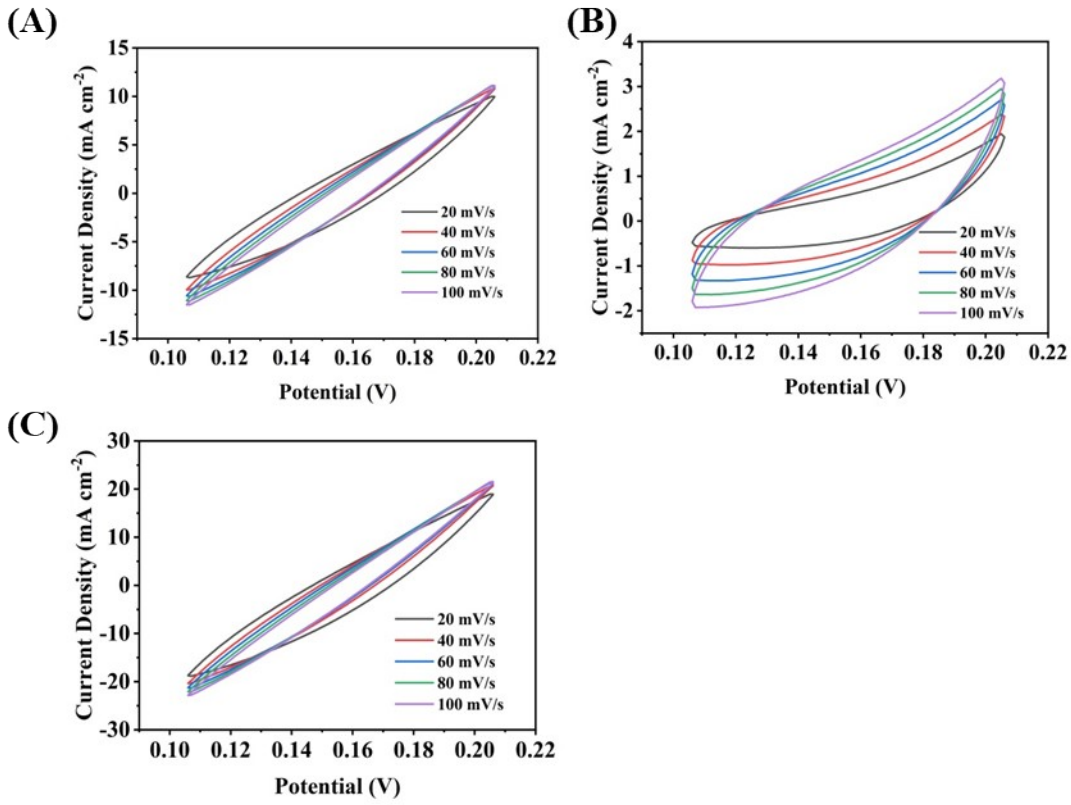

Figure S6. CV of (A) TiO_2@C, (B) CoCe-LDH and (C) $Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C at various scan rates for OER.

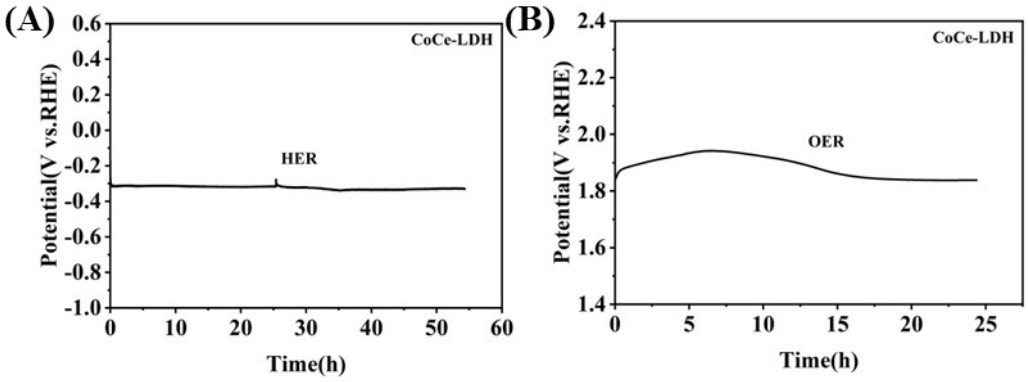

Figure S7. Chronopotentiometry measurement of CoCe-LDH of (A) HER and (B) OER performance.

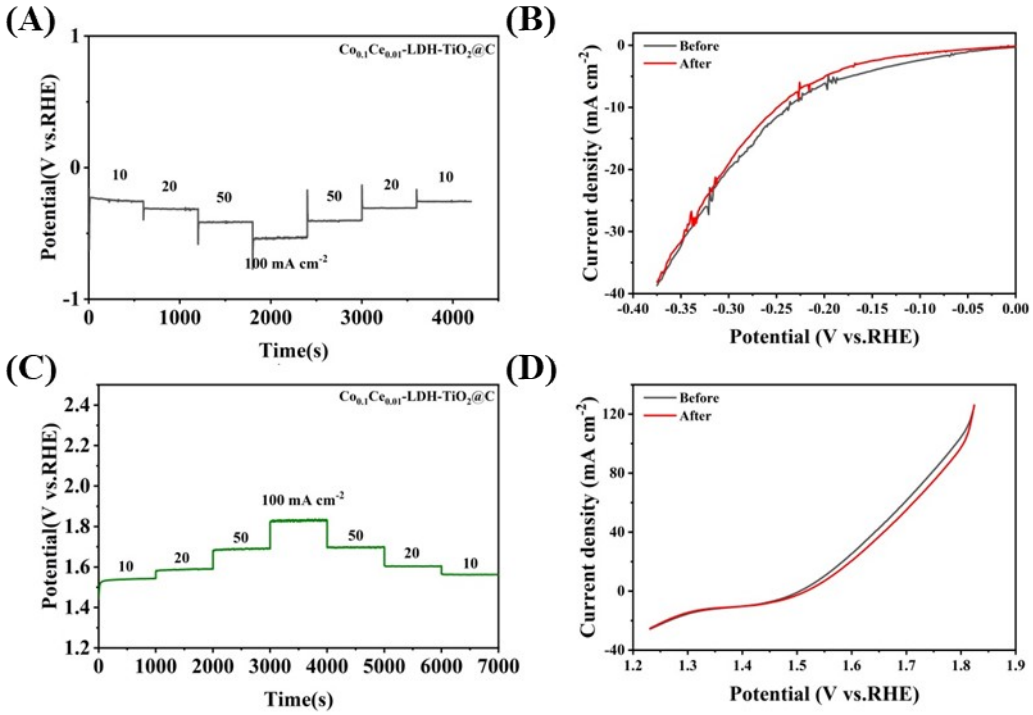

Figure S8. Stepped current stability test of $Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C for (A) HER and (C) OER, LSV curve comparison experiment before and after CV cycling of $Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C for (B) HER and (D) OER.

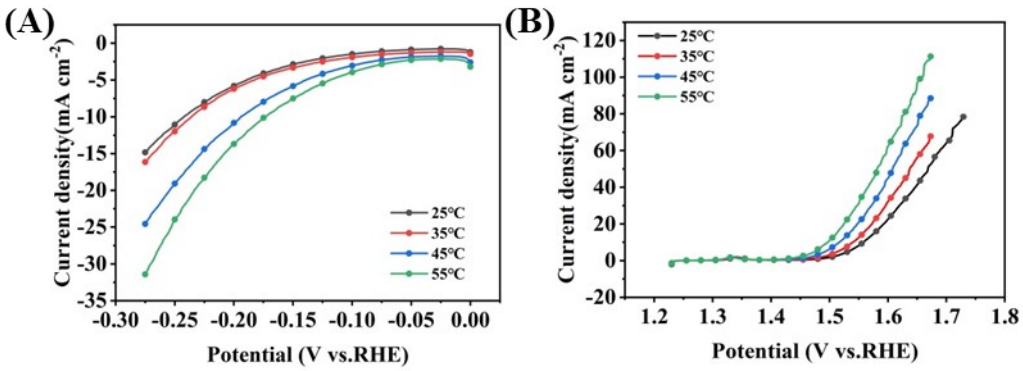

Figure S9. LSV polarization curves under different electrolyte temperatures of (A-B) CoCe-LDH.

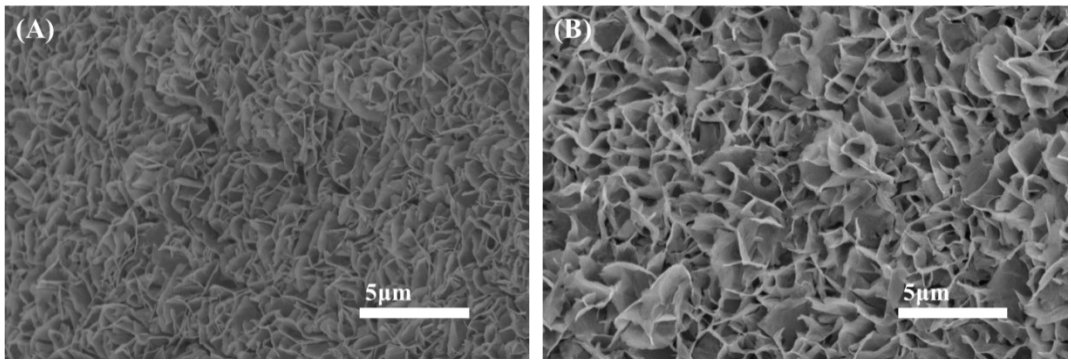

Figure S10. The SEM images of $Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C after (A) HER and (B) OER.

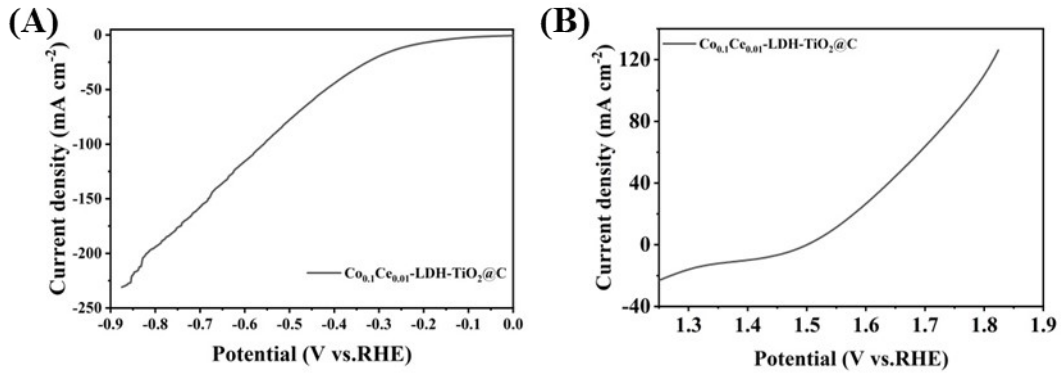

Figure S11. LSV polarization curves at high current density of $Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C for (A) HER and (B) OER.

Table S1. EIS parameters of TiO_2@C, CoCe-LDH and $Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C at different potentials for HER.

Samples	Potential (V vs. RHE)	Rct (Ω)	Rs (Ω)	CPE (mF)
TiO_2@C	-0.176 V	17.178	1.4995	0.85619
	-0.191 V	12.751	1.5959	0.87303
	-0.206 V	10.001	1.3743	0.82405
	-0.221 V	7.8108	1.1776	0.78498
	-0.236 V	6.7571	1.1382	0.75446
CoCe-LDH	-0.176 V	14.304	1.758	0.85989
	-0.191 V	10.968	1.8533	0.9105
	-0.206 V	9.0989	1.8542	0.92986
	-0.221 V	7.6343	1.8459	0.94465
	-0.236 V	6.4156	1.8839	0.94813
$Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C	-0.176 V	6.6029	1.4359	0.6716
	-0.191 V	6.1093	1.6649	0.74449
	-0.206 V	5.6102	1.4617	0.71749
	-0.221 V	4.494	1.4968	0.74299
	-0.236 V	3.5875	1.5288	0.7842

Table S2. EIS parameters of TiO_2@C, CoCe-LDH and $Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C at different potentials for OER.

Samples	Potential (V vs. RHE)	Rct (Ω)	Rs (Ω)	CPE (mF)
TiO_2@C	1.538 V	3.7844	1.0205	0.65756
	1.553 V	2.8462	1.0287	0.67259
	1.568 V	2.077	1.0125	0.75114
	1.583 V	1.4984	0.92555	0.7799
	1.598 V	1.3782	0.94059	0.81062
CoCe-LDH	1.538 V	3.3702	1.8442	0.82349
	1.553 V	2.4545	1.7925	0.82286
	1.568 V	1.7764	1.7896	0.83172
	1.583 V	1.3837	1.7826	0.83567
	1.598 V	1.1192	1.8153	0.84869
$Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C	1.538 V	2.1355	1.564	0.87563
	1.553 V	1.6479	1.5679	0.86365
	1.568 V	1.2684	1.5731	0.86883
	1.583 V	0.98091	1.5844	0.88856
	1.598 V	0.84179	1.569	0.8629

Table S3. Comparison of HER and OER performance for $Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C with other similar catalysts

Samples	η_{HER}	η_{OER}	C.D.	Ref.
$Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C	202 mV	268 mV	10 mA cm^{-2}	**This work**
Co-NiFe LDH		278 mV	10 mA cm^{-2}	[1]
FeOOH(Se)/IF		287 mV	10 mA cm^{-2}	[2]
Ni/$Ni(OH)_2$		270 mV	10 mA cm^{-2}	[3]
Fe-CoOOH/G		330 mV	10 mA cm^{-2}	[4]
ZnCo-C/CA-PANI@NF		338 mV	10 mA cm^{-2}	[5]
NiFe-LDH(POM)		287 mV	10 mA cm^{-2}	[6].
Ir–Co_3O_4		622 mV	10 mA cm^{-2}	[18]
$Co_{.85}Se/Co_9Se_8$		330 mV	10 mA cm^{-2}	[19]
NiFe-LDH/NF	210 mV		10 mA cm^{-2}	[7]
CoS/rGO	371 mV		10 mA cm^{-2}	[8].
$NiCo_2S_4$/NF	210 mV		10 mA cm^{-2}	[9]
Co-BDC/MoS_2	248 mV		10 mA cm^{-2}	[10]
$Ni(OH)_2/MoS_2$	227 mV		10 mA cm^{-2}	[11]
Dy-CuO	220 mV		10 mA cm^{-2}	[20]
CuO	243 mV		10 mA cm^{-2}	[20]
Co35Cr5Fe10Ni30Ti20	217 mV		10 mA cm^{-2}	[21]

η_{HER} is the overpotential of HER, η_{OER} is the overpotential of OER, C.D. is current density

Table S4. Comparison of the stability performance of $Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C with other catalysts for OER.

Samples	stability performance	Ref.
$Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C	100 h	**This work**
1T-$Mn_{0.8}Ir_{0.2}O_2$	75 h	[12]
NiFeMn-OOH/NF	90 h	[13]
CoP-Ni_2P@U-NCNTs/NF	48 h	[14]
Ni_2P@C	50 h	[15]
NiCoFeMoZn	50 h	[16]
PMOF-NiOOH-Ru_{20}/NF-A	50 h	[17]

Table S5. Comparison of the stability performance of $Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C with other catalysts for overall water splitting.

Samples	stability performance	Ref.
$Co_{0.1}Ce_{0.01}$-LDH-TiO_2@C	100 h	**This work**
IrO_2/MoS_2	70 h	[22]
NF-C/CoS/NiOOH	13.8 h	[23]
4N6Co-MoS_2	50 h	[24]
MS_2/RuS_2@G（M=Fe,Co,Ni）	20 h	[25]
$Ni_3Sb/FeSb_2$	50 h	[26]

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