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Two-Dimensional Stable and Ultrathin Cluster-based Metal-Organic Layers for Efficient Electrocatalytic Water Oxidation†

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Table S1. Crystallographic data and structure refinement details for **$Co_{0.6}Ni_{0.4}$-CMOLs** and **Co-**

CMOLs[1].

compound	$Co_{0.6}Ni_{0.4}$-CMOLs[this work]	Co-CMOLs[1]
formula	$C_{16}H_8Co_{1.2}Ni_{1.8}O_{10}$	$C_{16}H_8Co_3O_{10}$
formula weight	536.62	539.03
temperature (K)	293	293
crystal system	Monoclinic	Monoclinic
space group	$P\,2_1/c$	$P\,2_1/c$
a (Å)	11.7117 (14)	11.648 (2)
b (Å)	12.0990 (13)	12.091 (3)
c (Å)	11.7979 (13)	11.846 (3)
α (°)	90	90
β (°)	103.170 (12)	103.245 (4)
γ (°)	90	90
V (Å^3)	1627.8 (3)	1624.0 (6)
Z	4	4
D_x, g cm^{-3}	2.190	2.205
Mu, mm^{-1}	12.466	3.090
F(000)	1067.2	1068
GOF on F^2	1.001	0.9660
R_{int}	0.0734	0.0425
R_1, wR_2 [$I>2\sigma(I)$]	0.0866, 0.2141	0.0409, 0.1045
R_1, wR_2 [all data]	0.1264, 0.2550	0.0601, 0.1136
CCDC no.	2068005	913356

$R_1 = \Sigma||Fo|-|Fc||/\Sigma|Fo|$. $wR_2=[\Sigma w(Fo^2-Fc^2)^2/\Sigma w(Fo^2)^2]^{1/2}$.

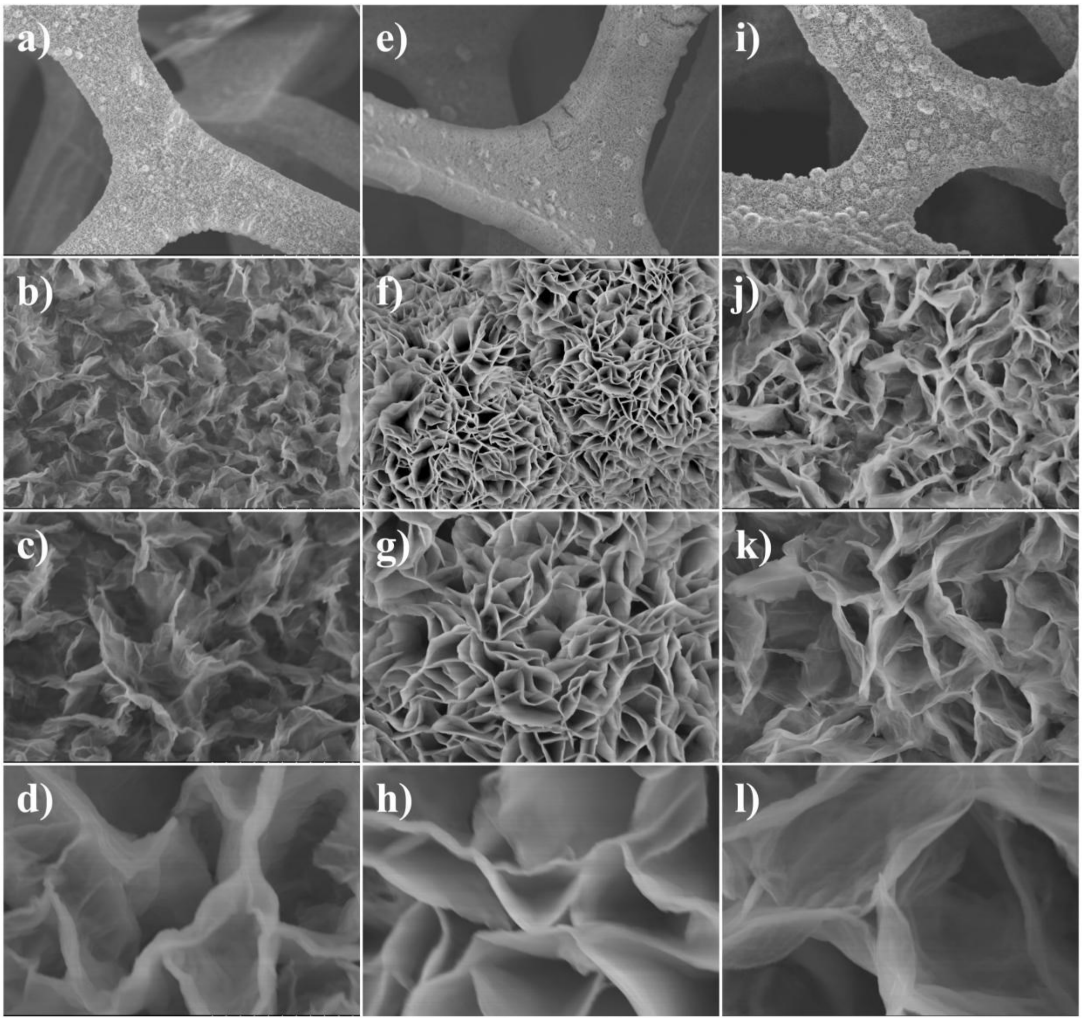

Fig. S1. SEM images of a) b), c), d) **Co-CMOLs/NF-KOH/1M**; e), f), g), h) **Ni-CMOLs/NF-KOH/1M** and i), j), k), l) **$Co_{0.6}Ni_{0.4}$-CMOLs/NF -KOH/1M**.

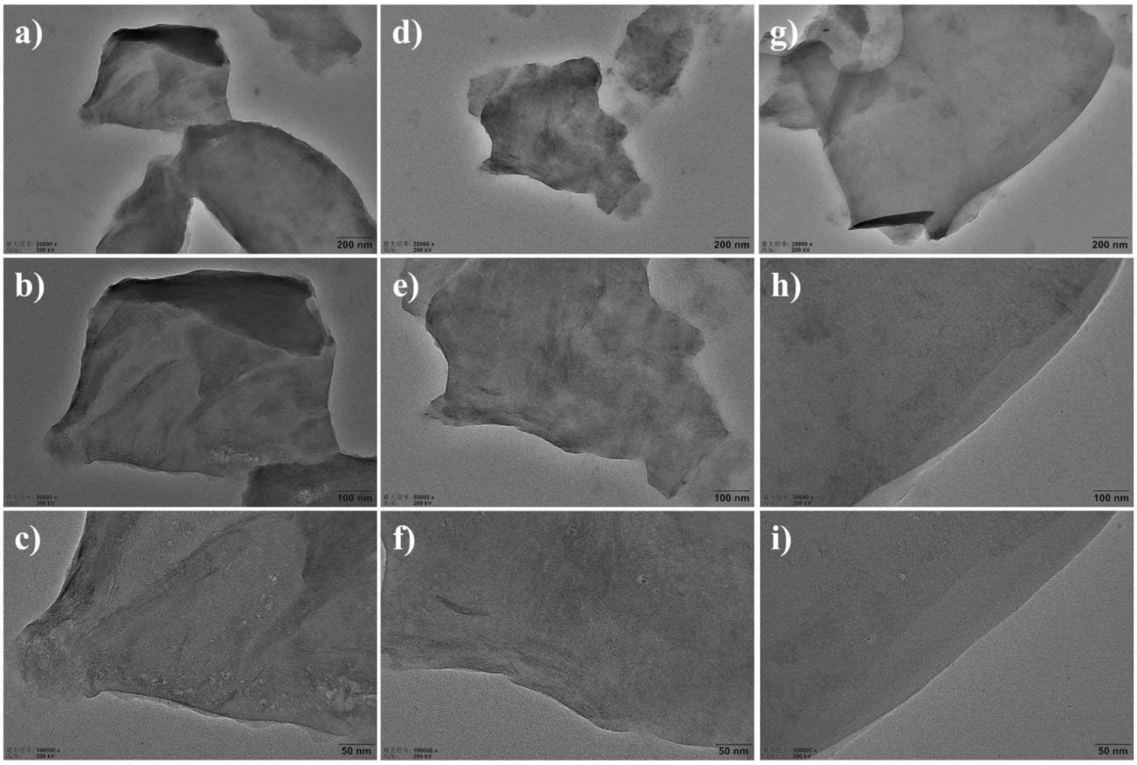

Fig. S2. TEM images of **$Co_{0.6}Ni_{0.4}$-CMOLs** (The **$Co_{0.6}Ni_{0.4}$-CMOLs** was removed from **$Co_{0.6}Ni_{0.4}$-CMOLs/NF** by ultrasonication)

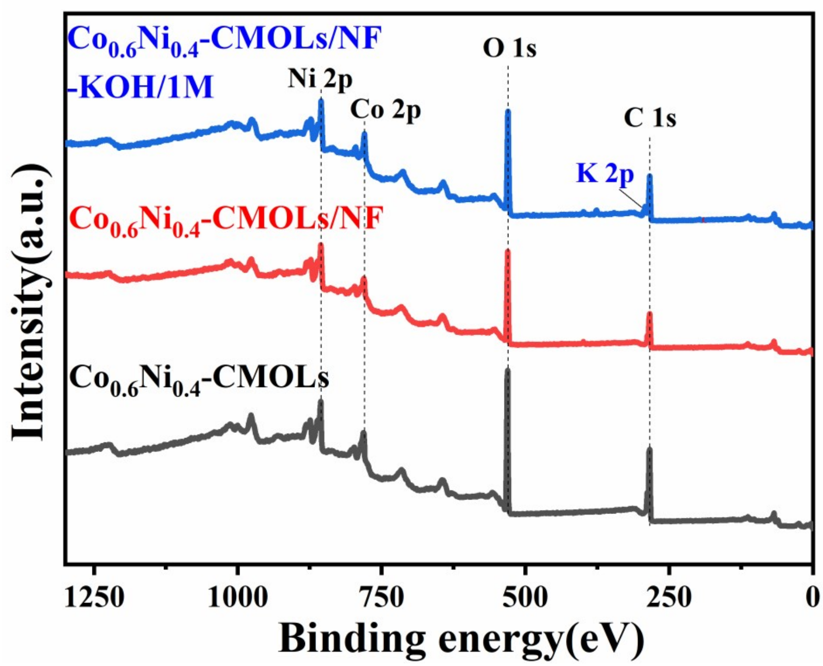

Fig. S3. XPS survey spectra for **$Co_{0.6}Ni_{0.4}$-CMOLs/NF-KOH/1M**, **$Co_{0.6}Ni_{0.4}$-CMOLs/NF** and **$Co_{0.6}Ni_{0.4}$-CMOLs**.

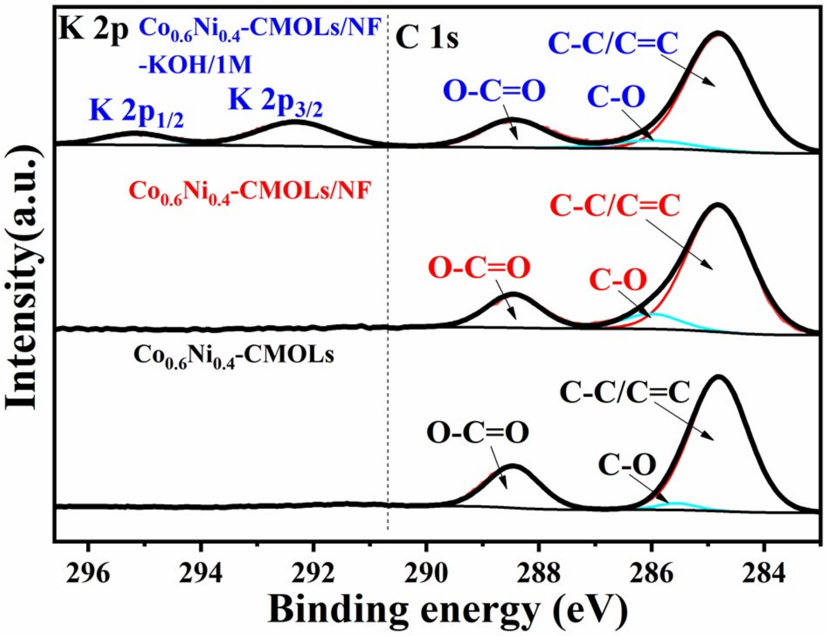

Fig. S4. The C 1s spectrum for **$Co_{0.6}Ni_{0.4}$-CMOLs/NF-KOH/1M**, **$Co_{0.6}Ni_{0.4}$-CMOLs/NF** and **$Co_{0.6}Ni_{0.4}$-CMOLs** (The K 2p shown in **$Co_{0.6}Ni_{0.4}$-CMOLs/NF-KOH/1M** is due to residual K^+).

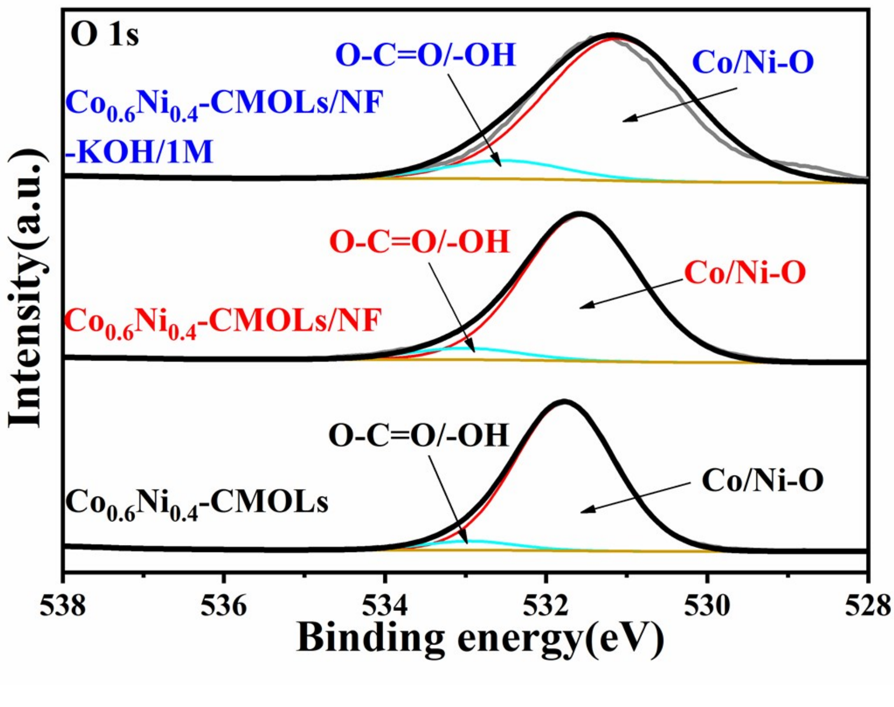

Fig. S5. The O 1s spectrum for **$Co_{0.6}Ni_{0.4}$-CMOLs/NF-KOH/1M**, **$Co_{0.6}Ni_{0.4}$-CMOLs/NF** and **$Co_{0.6}Ni_{0.4}$-CMOLs**.

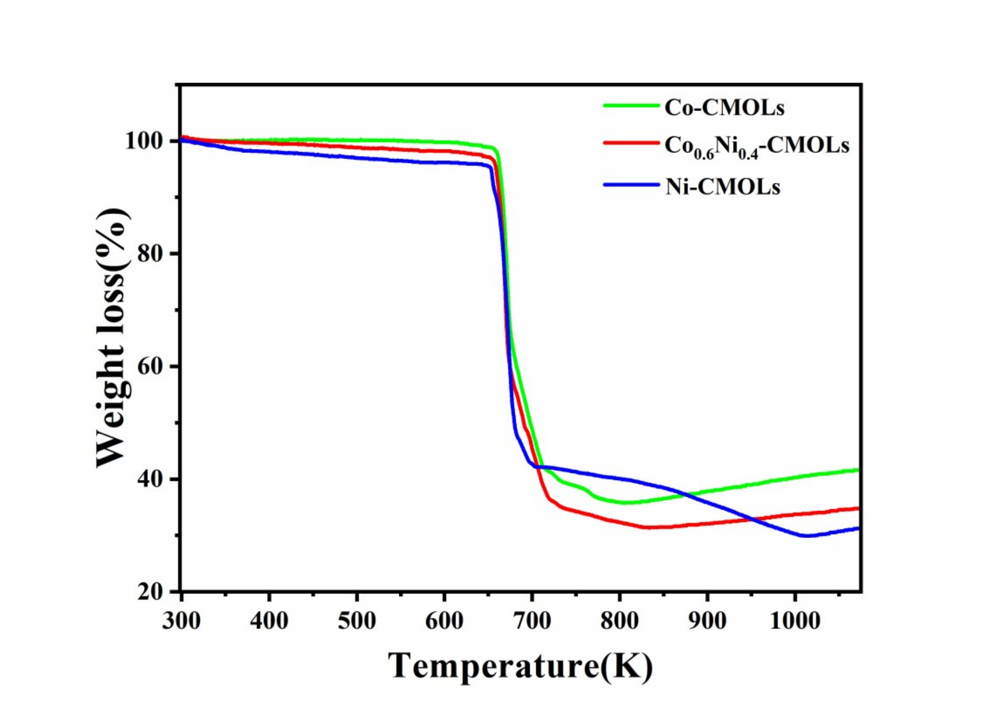

Fig.S6. TGA of **Co-CMOLs**, **$Co_{0.6}Ni_{0.4}$-CMOLs** and **Ni-CMOLs**.

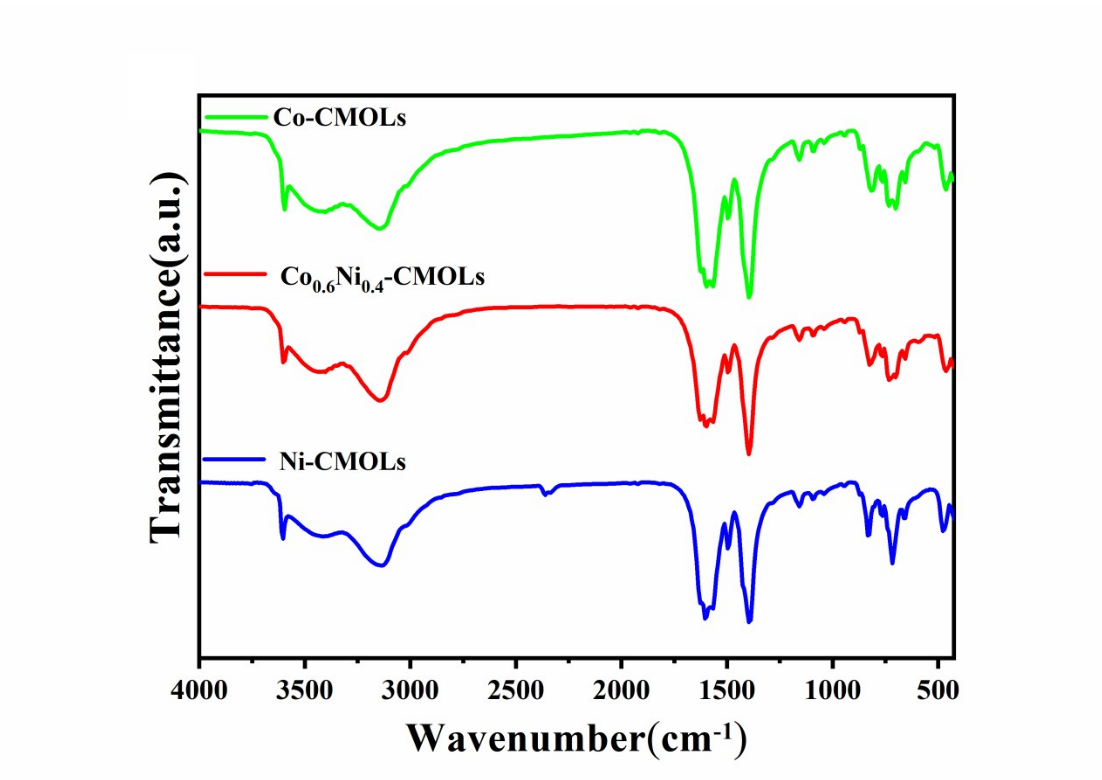

Fig. S7. IR of **Co-CMOLs**, **$Co_{0.6}Ni_{0.4}$-CMOLs** and **Ni-CMOLs**.

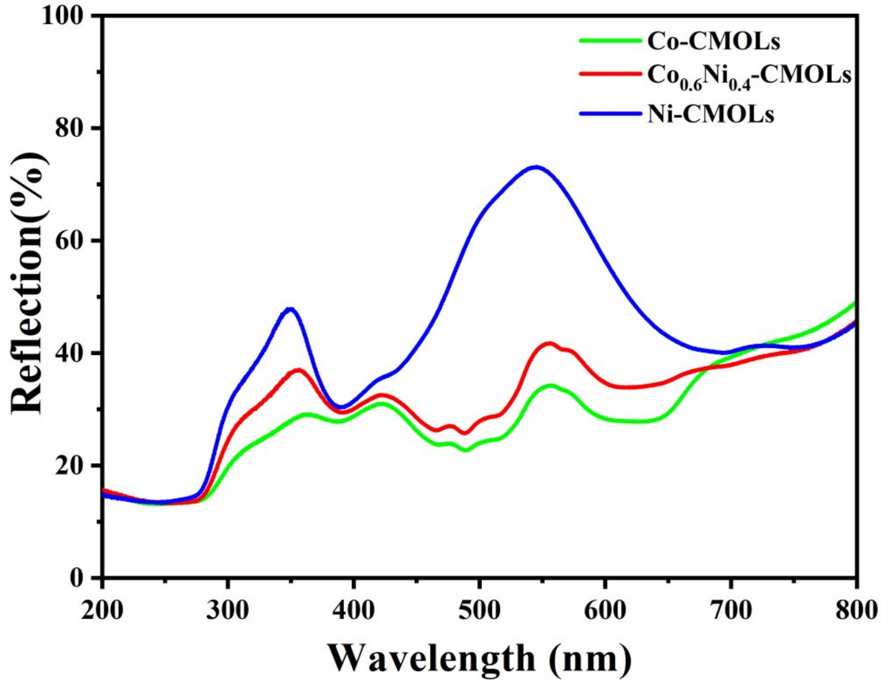

Fig. S8. UV-visible diffuse reflectance spectra of **Co-CMOLs**, **$Co_{0.6}Ni_{0.4}$-CMOLs** and **Ni-CMOLs**.

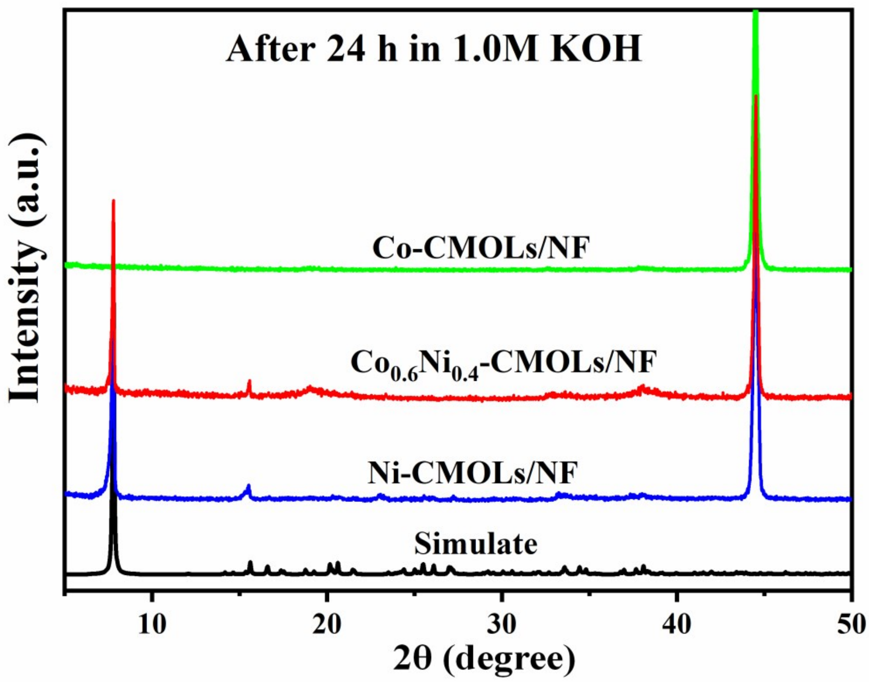

Fig. S9. PXRD patterns of three **CMOLs/NF** immersed in 1.0 M KOH for 24 h.

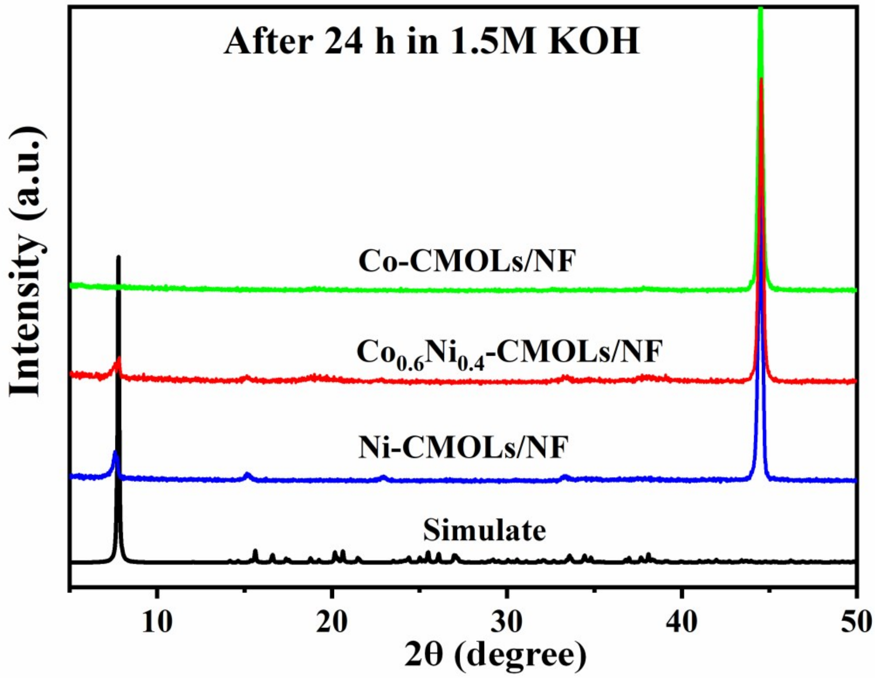

Fig. S10. PXRD patterns of three **CMOLs/NF** immersed in 1.5M KOH for 24 h.

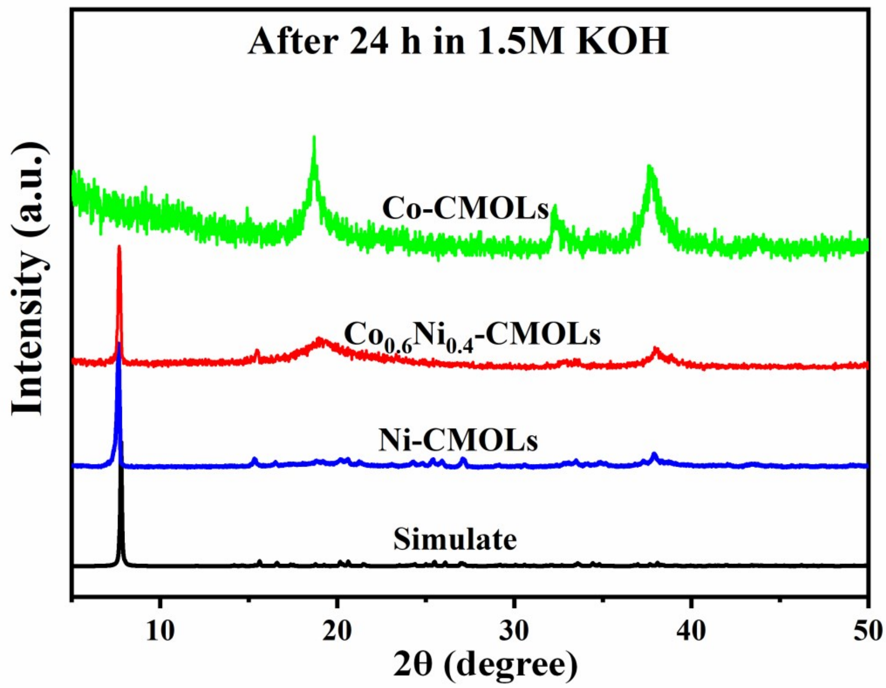

Fig. S11. PXRD patterns of three **CMOLs** immersed in 1.5M KOH for 24 h.

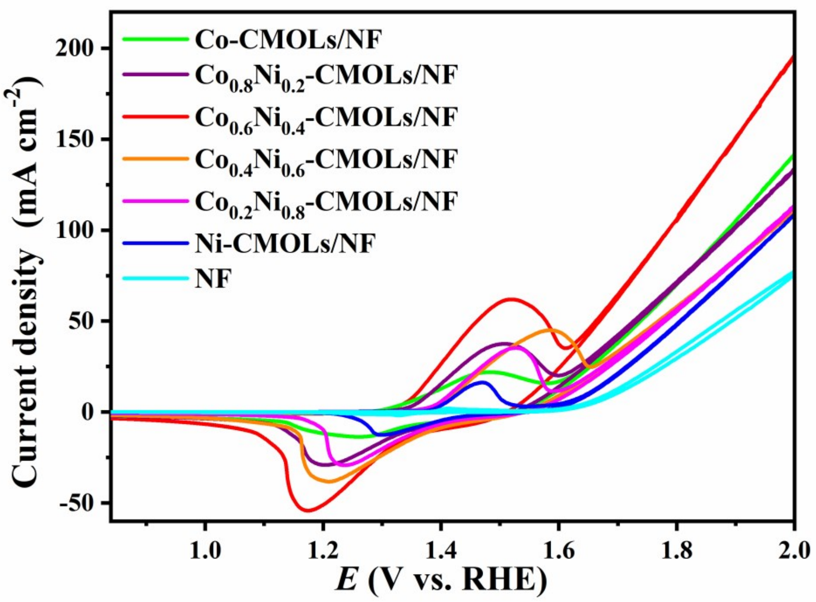

Fig. S12. CV curves of **Co-CMOLs/NF**, **Ni-CMOLs/NF**, **Co_xNi_{1-x}-CMOLs/NF** and blank **NF** in 1.0 M KOH for OER without iR correction.

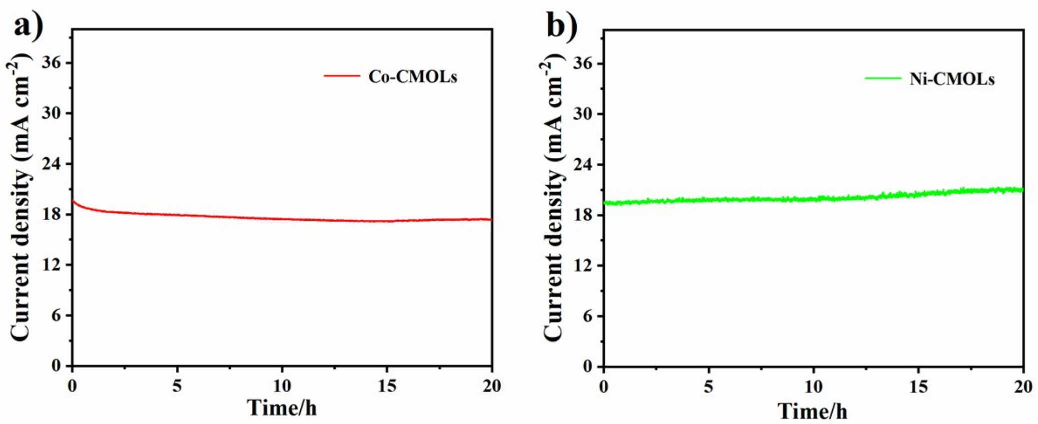

Fig. S13. a) Chronoamperometry acquired with **Co-CMOLs** under a constant potential for 20 h, b) Chronoamperometry acquired with **Ni-CMOLs** under a constant potential for 20 h.

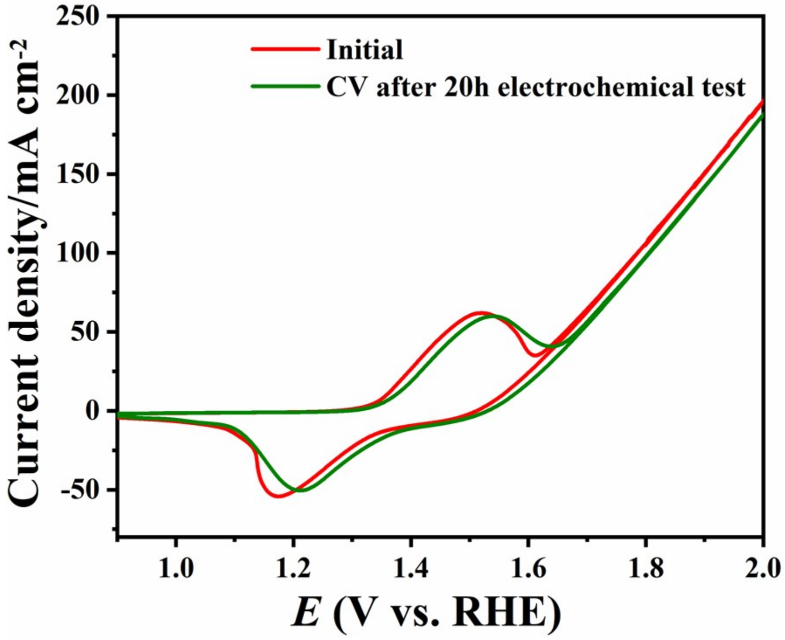

Fig. S14. Initial CV and CV after after 20 h electrochemical test.

Table S2: ICP analysis for **$Co_{0.6}Ni_{0.4}$-CMOLs**.

$Co_{0.6}Ni_{0.4}$-CMOLs	Sampling quality (g)	Volume (mL)	Coefficient of dilution	Instrument readings (mg/L)	Molar ratio
Co	0.0114	25	50	1.244	0.409
Ni	0.0114	25	50	1.791	0.591

Table S3: XPS high-resolution spectrum of **$Co_{0.6}Ni_{0.4}$-CMOLs**, **$Co_{0.6}Ni_{0.4}$-CMOLs/NF** and **$Co_{0.6}Ni_{0.4}$-CMOLs/NF-KOH/1M**.

$Co_{0.6}Ni_{0.4}$-CMOLs	FWHM eV	Area (P) CPS.eV	Atomic %
Co 2p	3.08	173031.74	40.51
Ni 2p	2.52	276214.97	59.49

$Co_{0.6}Ni_{0.4}$-CMOLs/NF	FWHM eV	Area (P) CPS.eV	Atomic %
Co 2p	4.04	111457.02	33.60
Ni 2p	2.90	239475.26	66.40

$Co_{0.6}Ni_{0.4}$-CMOLs/NF-KOH/1M	FWHM eV	Area (P) CPS.eV	Atomic %
Co 2p	3.48	109448.03	32.43
Ni 2p	2.78	247881.31	67.57

Table S4: OER performance of pristine MOFs comparison between recently reported studies and this work.[2]

MOFs	Over potential	Tafel slope	Electrolyte	Ref.
FeNi-DOBDC-3	270 ($\eta_{j=50}$)	49	1.0 M KOH	3
$Co_{0.6}Fe_{0.4}$-MOF-74	280	56	1.0 M KOH	4
Fe/$Ni_{2.4}$-MIL-53	244	48.7	1.0 M KOH	5
Fe/$Ni_{2.4}$/$Co_{0.4}$-MIL-53	219	53.5	1.0 M KOH	5
$(Ni_2Co_1)_{0.925}Fe_{0.075}$-MOF-NF	257	41.3	1.0 M KOH	6
HE-MOF-RT	245	54	1.0 M KOH	7
PCN-Fe_2Co-Fe_2Ni	271	67.7	1.0 M KOH	8
UTSA-16	408	77	1.0 M KOH	9
CUMSs-ZIF-67	320	53.7	1.0 M KOH	10
CoBDC-$Fc_{0.17}$	219	61	1.0 M KOH	11
NiFe-UMNs	260	30	1.0 M KOH	12
Ni-Fe-NS	221	56	1.0 M KOH	13
$Fe_{0.1}$-Ni-MOF/NF	264 ($\eta_{j=100}$)	69.8	1.0 M KOH	14
LM-160–12	274	46.7	1.0 M KOH	15
NiCo-BDC BMNSs	230	61	1.0 M KOH	16
CoFe-MOF-OH	310	41	1.0 M KOH	17
Fe_1Ni_2-BDC	260	35	1.0 M KOH	18
Co_3Fe-MOF	280	38	1.0 M KOH	19
$A_{2.7}$B-MOF-$FeCo_{1.6}$	288	39	1.0 M KOH	20
Co/Cu-MOF (3)	395	94	1.0 M KOH	21
$Co_{0.6}Ni_{0.4}$-CMOLs/NF	**320**	**95**	**1.0 M KOH**	**This work**

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