

## Supplementary Information

### **Ni<sub>0.33</sub>Co<sub>0.67</sub>MoS<sub>4</sub> Nanosheets as Bifunctional Electrolytic Water Catalyst for Overall Water Splitting**

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	2Theta/degree in XRD pattern	d(nm) by calculation	2Theta/degree in JCPDS	d(nm) in JCPDS
NiCo-(MoO <sub>4</sub> ) <sup>2-</sup> LDH	34.01 59.79	0.264 0.155	34.742 60.457	0.258 0.153
$\text{Ni}_{0.33}\text{Co}_{0.67}\text{MoS}_4$	13.2	0.671	13.243	0.668
	20.4	0.4354	20.884	0.425
	22.3	0.3987	22.49	0.395
	26.9	0.3315	26.914	0.331
	29.3	0.3048	29.355	0.304
	30.6	0.2922	30.698	0.291
	32.17	0.2783	32.778	0.273
	35.7	0.2515	35.787	0.2507

Table S1. The calculated lattice parameters of NiCo-(MoO<sub>4</sub>)<sup>2-</sup> LDH and  $\text{Ni}_{0.33}\text{Co}_{0.67}\text{MoS}_4$ .

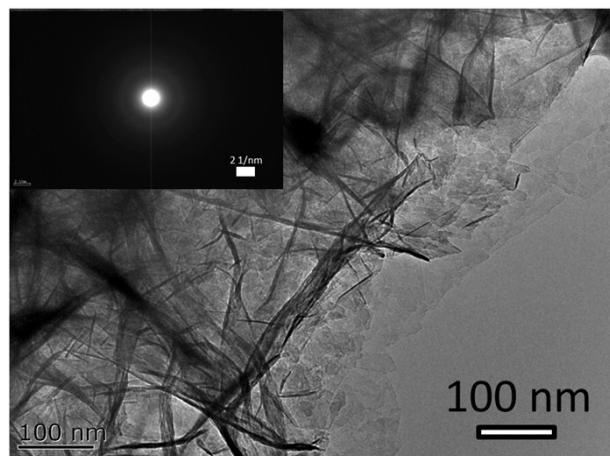


Figure S1. TEM image of NiCo-(MoO<sub>4</sub>)<sup>2-</sup> LDH and inset image from SAED.

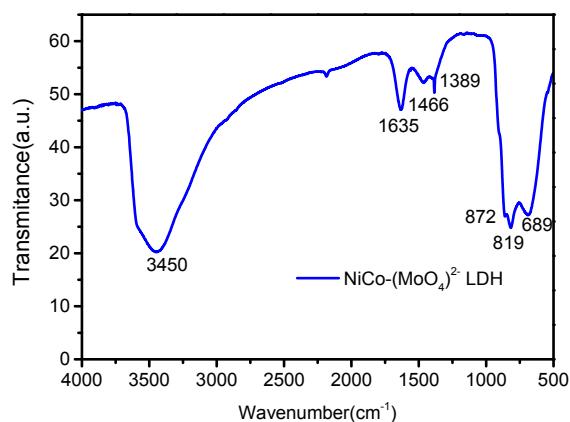


Figure S2. Infrared spectrum of NiCo-(MoO<sub>4</sub>)<sup>2-</sup> LDH.

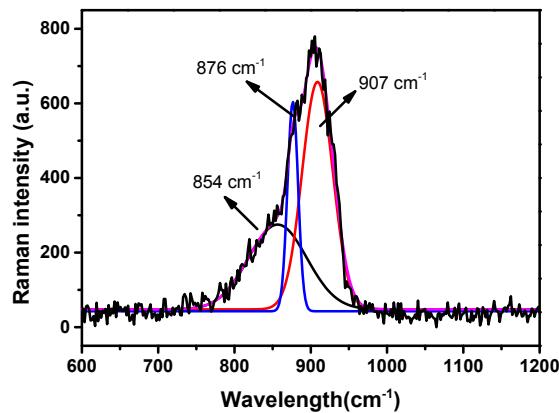


Figure S3. Raman spectrum of NiCo-(MoO<sub>4</sub>)<sup>2-</sup> LDH.

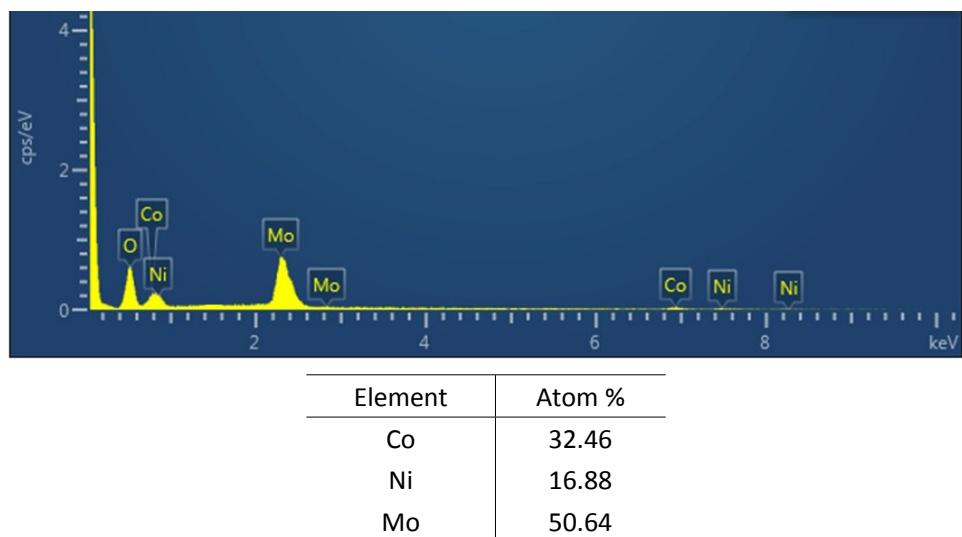
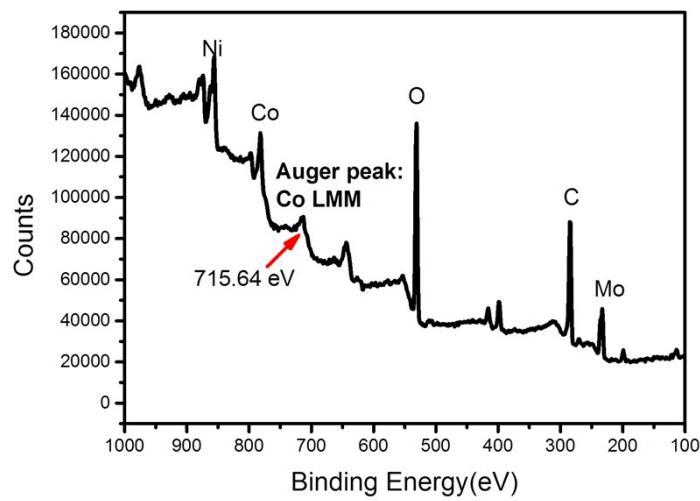


Figure S4. EDS spectrum of NiCo-(MoO<sub>4</sub>)<sup>2-</sup> LDH/CFC, and the atomic content of Ni, Co and Mo elements.

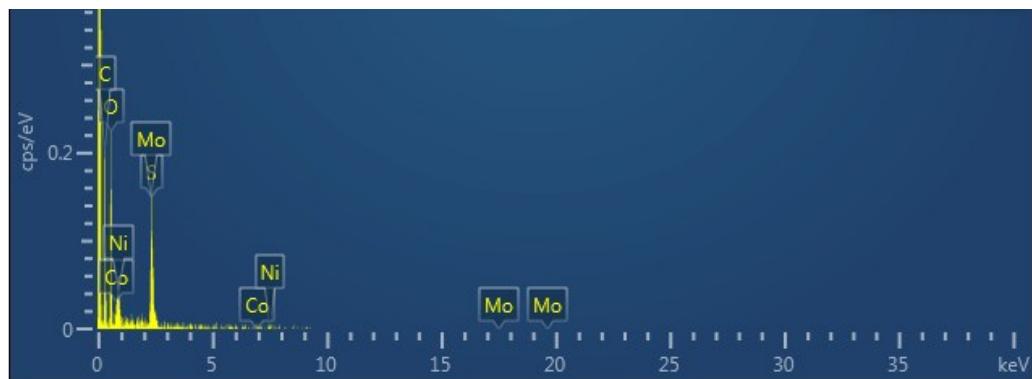
Sample	Element	Content ( $\mu\text{g/mL}$ )	Atom %
NiCo-(MoO <sub>4</sub> ) <sup>2-</sup> LDH	Co	6.255	34.1
	Ni	2.993	16.37
	Mo	14.835	49.53
Ni <sub>0.33</sub> Co <sub>0.67</sub> MoS <sub>4</sub> 4	Co	4.104	11.3
	Ni	1.962	5.4
	Mo	9.725	16.5
	S	13.15	66.8

Table S2. ICP-MS of Ni, Co, and Mo from NiCo-(MoO<sub>4</sub>)<sup>2-</sup> LDH and Ni<sub>0.33</sub>Co<sub>0.67</sub>MoS<sub>4</sub>.



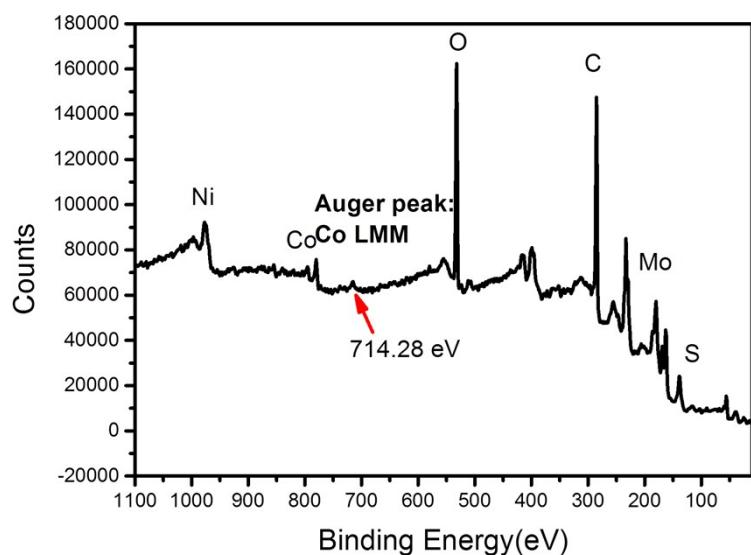
Element	Atom %
Co	8.37
Ni	4.14
Mo	12.54
C	74.95

Figure S5. Full XPS spectrum of NiCo-(MoO<sub>4</sub>)<sup>2-</sup> LDH, and the atomic content of Ni, Co and Mo elements from XPS.



Element	Atom %
Co	11.17
Ni	5.49
Mo	16.75
S	66.55

Figure S6. EDS spectrum of Ni<sub>0.33</sub>Co<sub>0.67</sub>MoS<sub>4</sub>/CFC, and the atomic content of Ni, Co, Mo and S elements.



Element	Atom %
Co	11.41
Ni	5.36
Mo	16.78
S	66.43

Figure S7. Full XPS spectrum of  $\text{Ni}_{0.33}\text{Co}_{0.67}\text{MoS}_4/\text{CFC}$ , and the atomic content of Ni, Co, Mo and S elements from XPS.

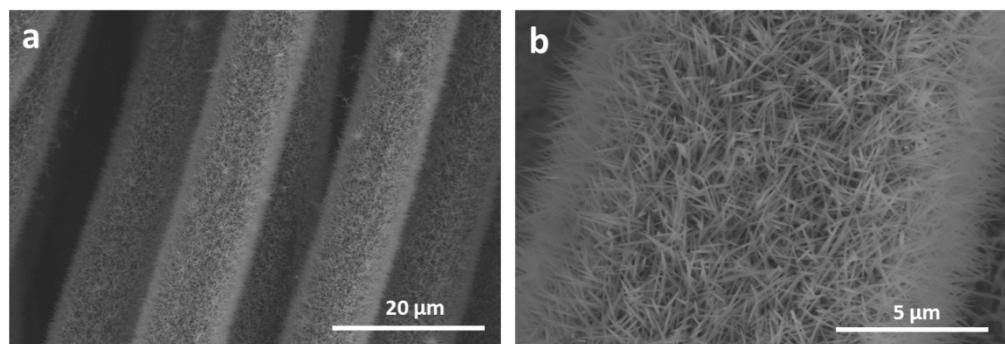


Figure S8. SEM images of  $\text{NiCo}_2(\text{CO}_3)_{1.5}(\text{OH})_3$  NW.

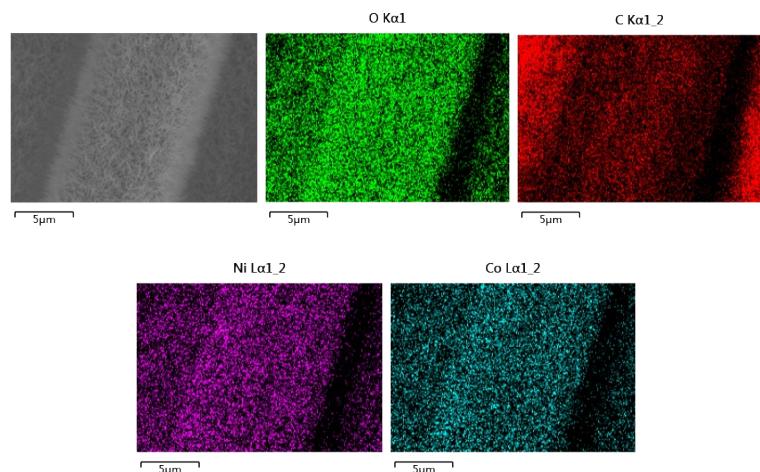


Figure S9. EDS mapping spectrum of  $\text{NiCo}_2(\text{CO}_3)_{1.5}(\text{OH})_3$  NW.

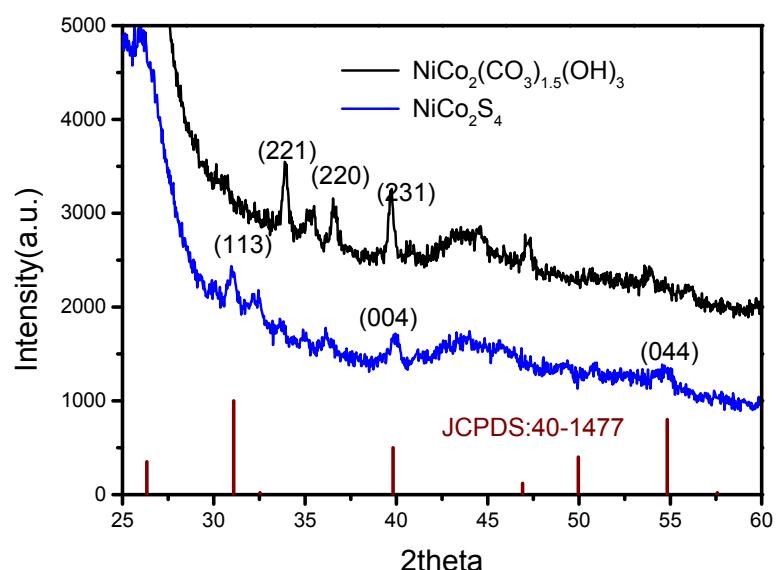


Figure S10. XRD patterns of  $\text{NiCo}_2(\text{CO}_3)_{1.5}(\text{OH})_3$  NW and  $\text{NiCo}_2\text{S}_4$  NW.

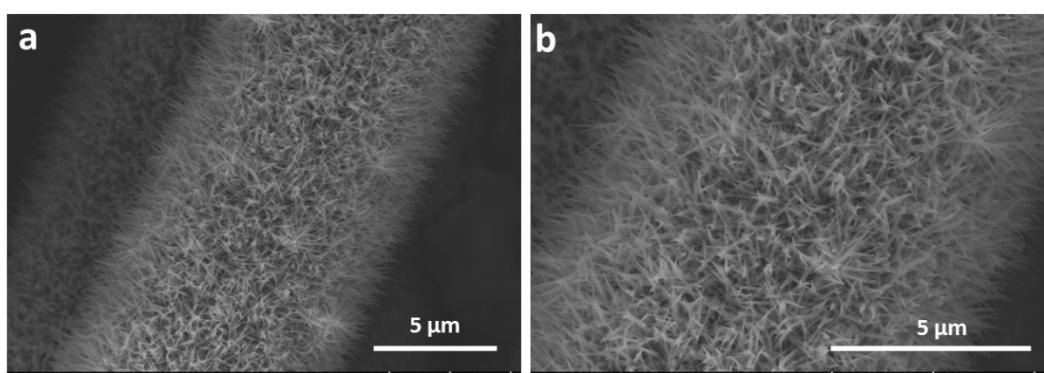


Figure S11. SEM images of  $\text{NiCo}_2\text{S}_4$  NW

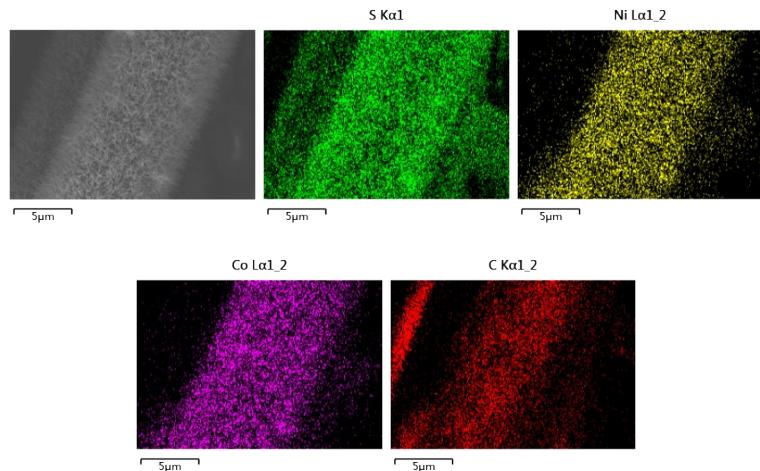


Figure S12. EDS mapping spectrum of  $\text{NiCo}_2\text{S}_4$  NW.

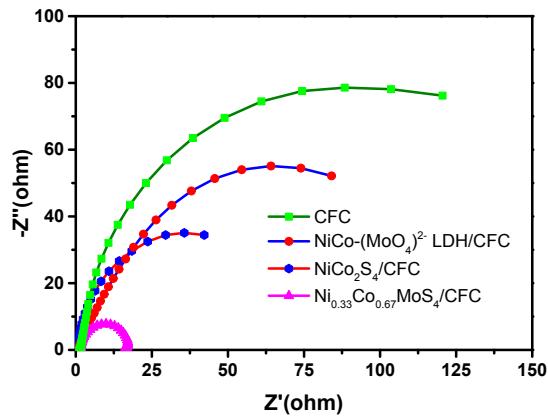


Figure S13. Electrochemical impedance spectra of  $\text{NiCo}-(\text{MoO}_4)^{2-}$  LDH/CFC,  $\text{Ni}_{0.33}\text{Co}_{0.67}\text{MoS}_4/\text{CFC}$ ,  $\text{NiCo}_2\text{S}_4/\text{CFC}$  and bare CFC were recorded at a potential of 0.5 V, sweeping the frequency from 100 kHz to 0.1Hz.

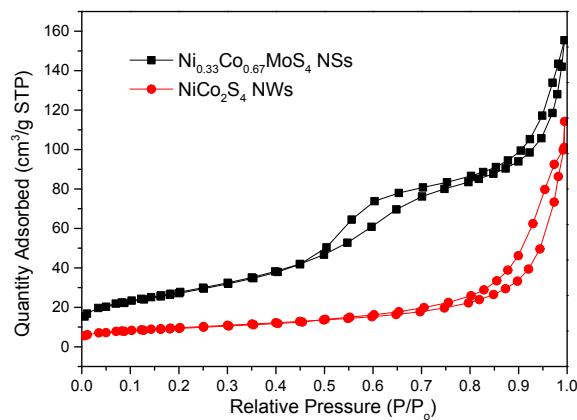


Figure S14.  $\text{N}_2$  isotherms of  $\text{Ni}_{0.33}\text{Co}_{0.67}\text{MoS}_4$  nanosheets and  $\text{NiCo}_2\text{S}_4$  nanowires stripped from CFC.

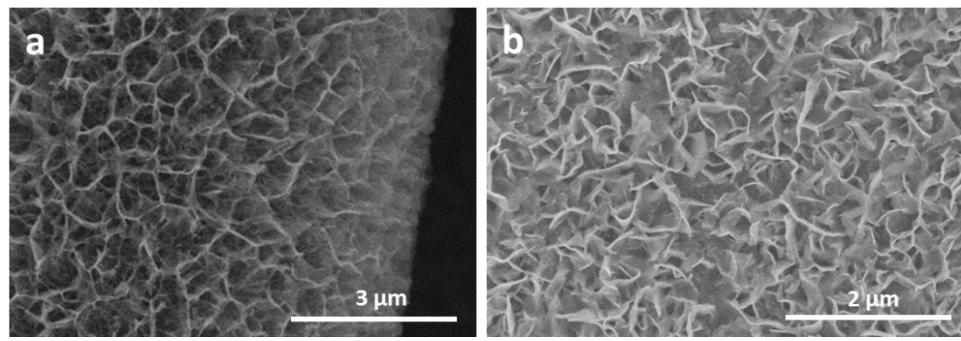


Figure S15. SEM images of Ni<sub>0.33</sub>Co<sub>0.67</sub>MoS<sub>4</sub>/CFC before and after electrochemical testing.

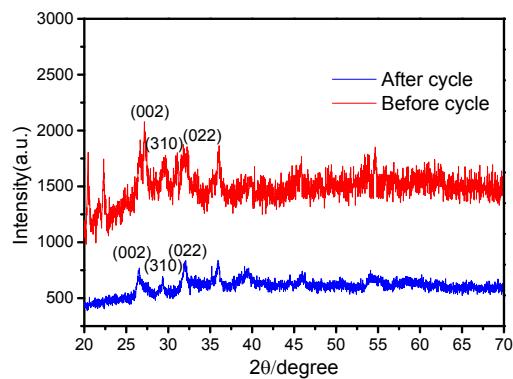


Figure S16. XRD patterns of Ni<sub>0.33</sub>Co<sub>0.67</sub>MoS<sub>4</sub>/CFC before and after electrochemical testing.

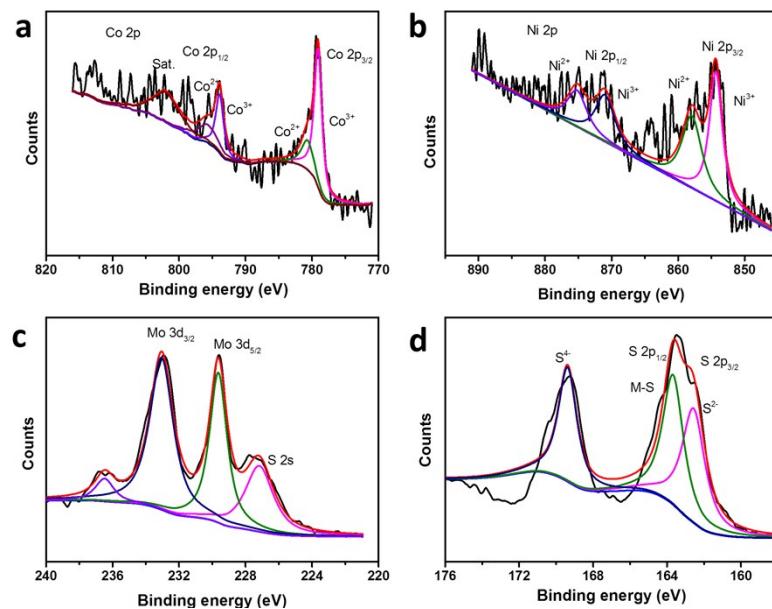


Figure S17 XPS spectra of Ni<sub>0.33</sub>Co<sub>0.67</sub>MoS<sub>4</sub>/CFC after electrochemical testing.

Catalyst	Support	Overall voltage(V) @10 mA cm <sup>-2</sup>	Reference
NiS	Ni foam	1.64 V	1
NiCo <sub>2</sub> S <sub>4</sub>	Carbon Cloth	1.68 V	2
Ni <sub>2</sub> P NPs	Ni foam	1.63 V	3
NiCo <sub>2</sub> S <sub>4</sub> NW	Ni foam	1.63 V	4
Ni-Co-P HNBs	Ni foam	1.62 V	5
Ni <sub>0.33</sub> Co <sub>0.67</sub> S <sub>2</sub> NW	Ti foil	1.65 V(5 mA cm <sup>-2</sup> )	6
NiCo <sub>2</sub> O <sub>4</sub> hollow microcuboids	Ni foam	1.65	7
Ni <sub>0.33</sub> Co <sub>0.67</sub> MoS <sub>4</sub>	Carbon Cloth	1.55 V	This work

Table S3. Comparison cell voltage of Ni<sub>0.33</sub>Co<sub>0.67</sub>MoS<sub>4</sub>/CFC//Ni<sub>0.33</sub>Co<sub>0.67</sub>MoS<sub>4</sub>/CFC with other bifunctional electrocatalyst in 1 M KOH solution.

### Reference

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