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## SUPPORTING INFORMATION



Fig. S1 XRD patterns of products at different reaction times.



Fig. S2 (a) SEM images of CoMoO<sub>4</sub>; (b) HRTEM images of CoMoO<sub>4</sub> inset SAED



Fig. S3 Roman spectra of CoMoO<sub>4</sub> and CoMoO<sub>4</sub>-Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub>



Fig. S4 SEM images of different hydrogen treatment time (a) 2h and (b) 6h.



Fig. S5 TEM images of hydrogen treatment for 6 hours



**Fig. S6** EDLC Cyclic voltammetry (CV) tests at different scan rates in non-Faradaic regions for (a) CoMoO<sub>4</sub> (b) CoMoO<sub>4</sub>-Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub> (c) Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub>



**Fig. S7** (a) CV curves for CoMoO<sub>4</sub>, CoMoO<sub>4</sub>-Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub> and Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub> recorded in 1.0 M PBS (PH=7) at scan rate of 50 mV s<sup>-1</sup>. (b) Comparison of the TOFs of CoMoO<sub>4</sub>, CoMoO<sub>4</sub>-Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub> and Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub>.

The absolute component of the voltammetric charge (cathode and anode) can be obtained from the CV of Fig. S7a. We assume it is one electronic redox process that divides this absolute charge by two, the number of active sites n (mol) can be calculated by the following formula:

n=Q/2FThe TOF (s<sup>-1</sup>) is calculated with the following equation : TOF=I/2Fn

- F: Faraday constant 96485 C/mol
- I: Current during the LSV measurement.

n: Number of active sites (mol)



**Fig. S8** (a) Chronoamperometry curves of CoMoO<sub>4</sub>, CoMoO<sub>4</sub>-Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub>, Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub> and Pt/C at 10 mA  $\cdot$  cm<sup>-2</sup> for 30 h.(b) Chronoamperometry curves of CoMoO<sub>4</sub>, CoMoO<sub>4</sub>-Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub> and Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub> at 10 mA  $\cdot$  cm<sup>-2</sup> for 300 h.

As shown in Fig. S8a, the potential of the CoMoO<sub>4</sub>, CoMoO<sub>4</sub>-Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub>,

 $Co_2Mo_3O_8$  and commercial Pt/C increased by 15 mV,11 mV, 25 mV and 8 mV at 10 mA·cm<sup>-1</sup> for 30h, respectively.

In the 300 h long-term stability test (Fig. S8b), the potentials of the three samples fluctuated in a small range. The potential fluctuation ranges of CoMoO<sub>4</sub>, CoMoO<sub>4</sub>-Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub>, Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub> were approximately 30 mV,21 mV and 38 mV at 10 mA $\cdot$ cm<sup>-1</sup> for 300 h, respectively.



**Fig. S9** (a) LSV curves of CoMoO<sub>4</sub>, CoMoO<sub>4</sub>-Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub> and Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub>. (b) Tafel plots of CoMoO<sub>4</sub>, CoMoO<sub>4</sub>-Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub>, Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub>. (c) LSV curves of CoMoO<sub>4</sub>-Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub> for overall water splitting in a two-electrode configuration. (d) Chronoamperometry curve of CoMoO<sub>4</sub>-Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub> for overall water splitting at 10 mA cm<sup>-2</sup>(inset of digital image of two-electrode water splitting.).



Fig. S10 SEM images of CoMoO<sub>4</sub>-Co<sub>2</sub>Mo<sub>3</sub>O<sub>8</sub> after stability test at different magnifications.