## **Supporting Information**

Electrodeposition of  $Ni_3Se_2/MoSe_x$  as a bifunctional electrocatalyst towards highly-efficient overall water splitting

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Fig. S1. SEM images of  $Ni_3Se_2(a-c)$  and  $MoSe_x$  (d-f).



Fig. S2. EDX and ICP results of  $Ni_3Se_2$  and  $Ni_3Se_2$ /MoSe<sub>x</sub>.



Fig. S3. XPS survey spectrum of  $Ni_3Se_2/MoSe_x$ . The inset shows the determined Ni, Mo and Se content in Ni3Se2/MoSe.



Fig. S4. CTTs curves of Ni<sub>3</sub>Se<sub>2</sub>, MoSe<sub>x</sub> and Ni<sub>3</sub>Se<sub>2</sub>/MoSe<sub>x</sub> at -0.8V versus Ag/AgCl.



Fig. S5. The cyclic voltammetry (CV) curves of  $Ni_3Se_2$  and  $Ni_3Se_2/MoSe_x$  in the range of -0.8 and -0.9 V (vs. Ag/AgCl) at different scan rates.



Fig. S6. The Chronopotentiometry measurement (potential vs time) of  $Ni_3Se_2/MoSe_x$  at -10 mA cm<sup>-2</sup>.



Fig. S7. The Chronopotentiometry measurement (potential vs time) of  $Ni_3Se_2/MoSe_x$  at 10 mA cm<sup>-2</sup>.

Electrocatalyst	Overpotential (mV) at 10 mA/cm <sup>2</sup>	Tafel slope (mV/dec)	Electrolyte	References
Ni <sub>3</sub> Se <sub>2</sub> /MoSe <sub>x</sub>	82	86.6	1.0 M KOH	Present work
Co-MoS <sub>2</sub> /rGO <sup>1</sup>	147	49.5	1.0 M KOH	Ref. 1
ZnMoO <sub>4</sub> /3D- AWC <sup>2</sup>	124	54	1.0 M KOH	Ref. 2
$Ni_5P_4$ - $Ru^3$	123	56.7	1.0 M KOH	Ref. 3
NiFeO <sub>x</sub> /CFP <sup>4</sup>	88	150.2	1.0 M KOH	Ref. 4
Mo <sub>x</sub> C-Ni@NCV <sup>5</sup>	126	93	1.0 M KOH	Ref. 5
NiSe <sup>6</sup>	96	120	1.0 M KOH	Ref. 6
NiMoN <sup>7</sup>	109	95	1.0 M KOH	Ref. 7
NiSe <sup>8</sup>	177	58.2	1.0 M KOH	Ref. 8
Ag <sub>2</sub> S/MoS <sub>2</sub> /RGO <sup>9</sup>	190	56	1.0 M KOH	Ref. 9
Co(OH)2@Ni10	96	104.2	1.0 M KOH	Ref. 10

 Table S1. The HER activities of Ni-/Mo-involved electrocatalysts.

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