

## Supporting Information

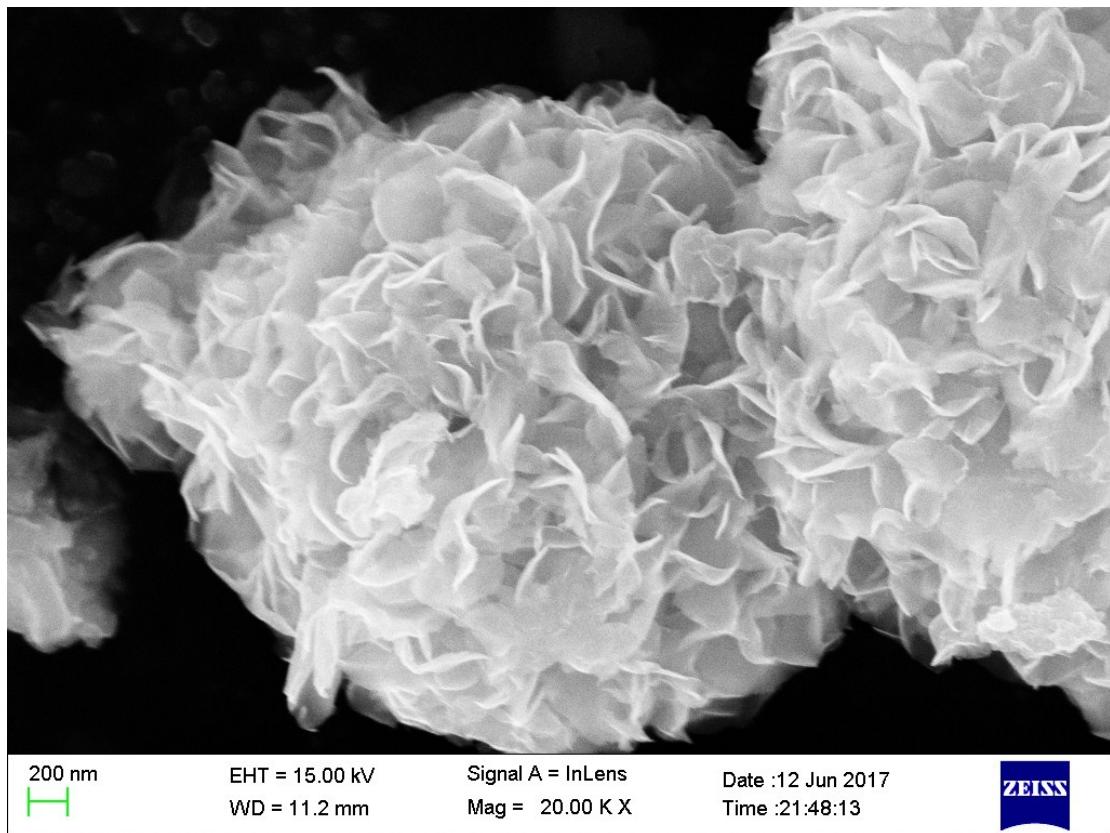
Surface Reorganization Engineering on N Dopants MoS<sub>2</sub> by In-situ  
Electrochemically Oxidation Activated for Efficient Oxygen Evolution  
Reaction

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200 nm  

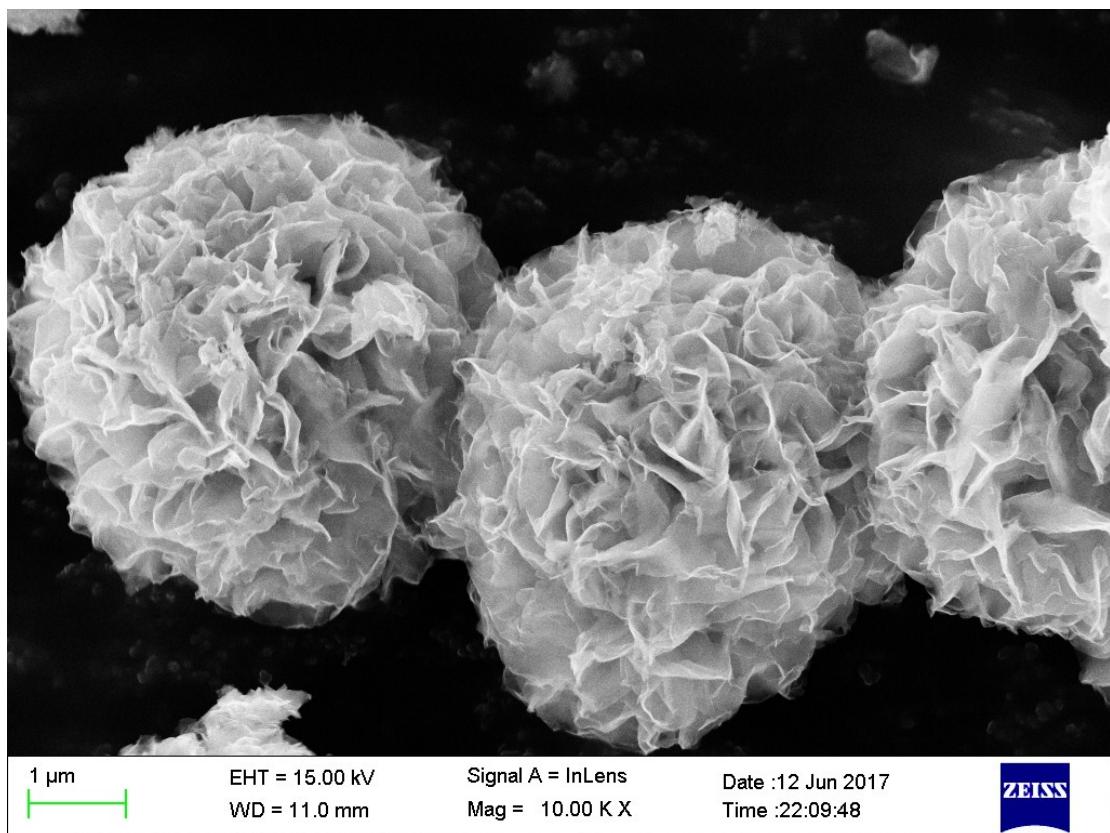

EHT = 15.00 kV  
WD = 11.2 mm

Signal A = InLens  
Mag = 20.00 K X

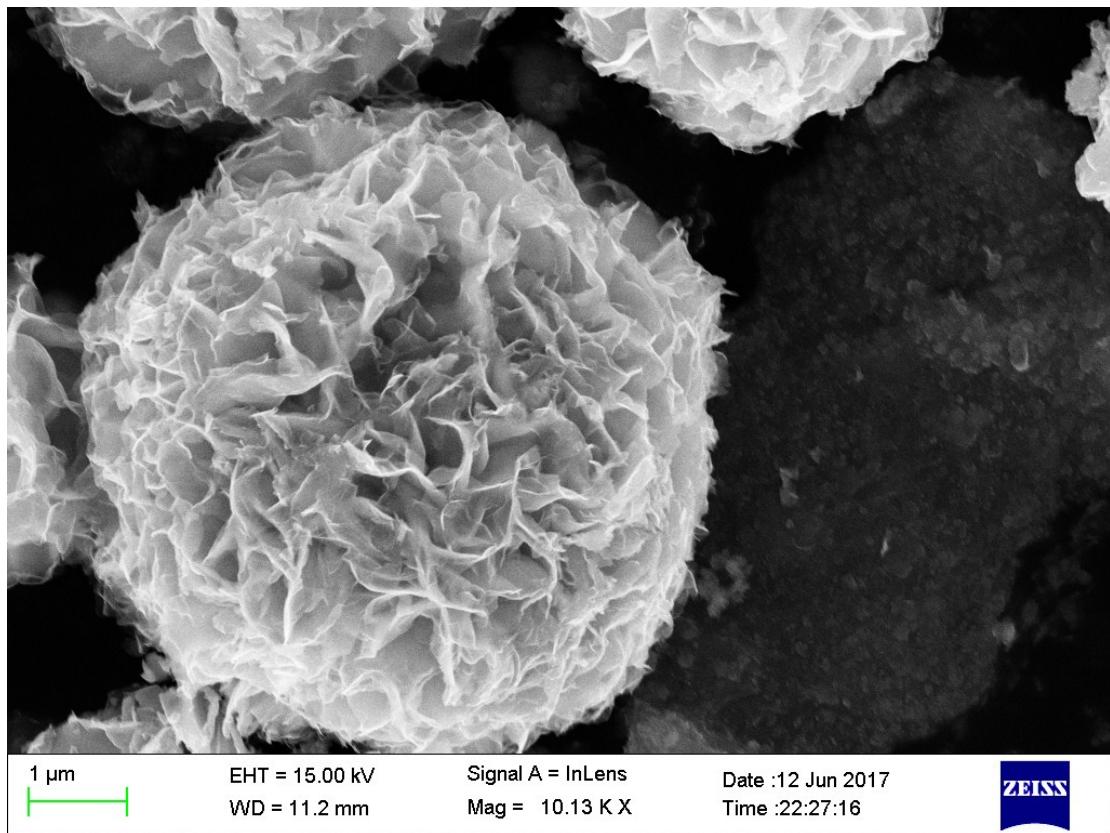
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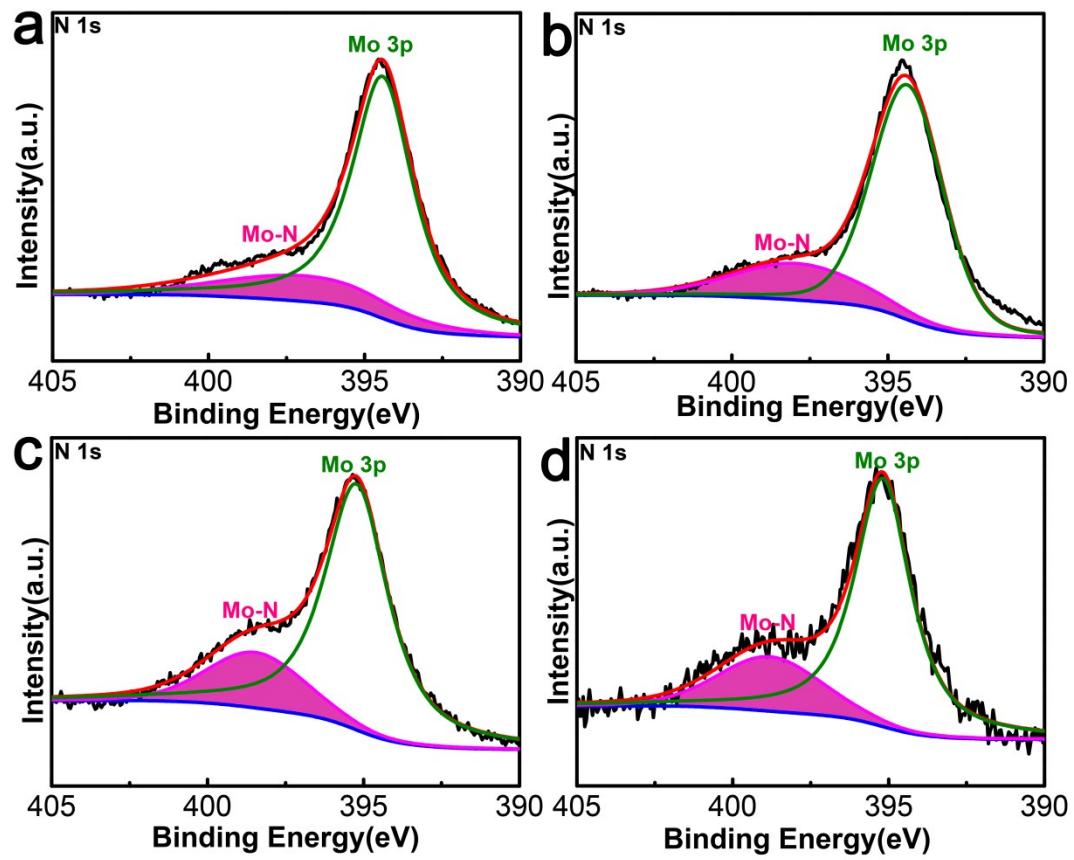
**Fig. S1 The SEM of N0-MoS<sub>2</sub>**



**Fig. S2 The SEM of N1-MoS<sub>2</sub>**



**Fig. S3 The SEM of N<sub>2</sub>-MoS<sub>2</sub>**



**Fig. S4** The XPS spectra of N 1s orbitals for N0-MoS<sub>2</sub>,N1-MoS<sub>2</sub>,N2-MoS<sub>2</sub> and N3-MoS<sub>2</sub>

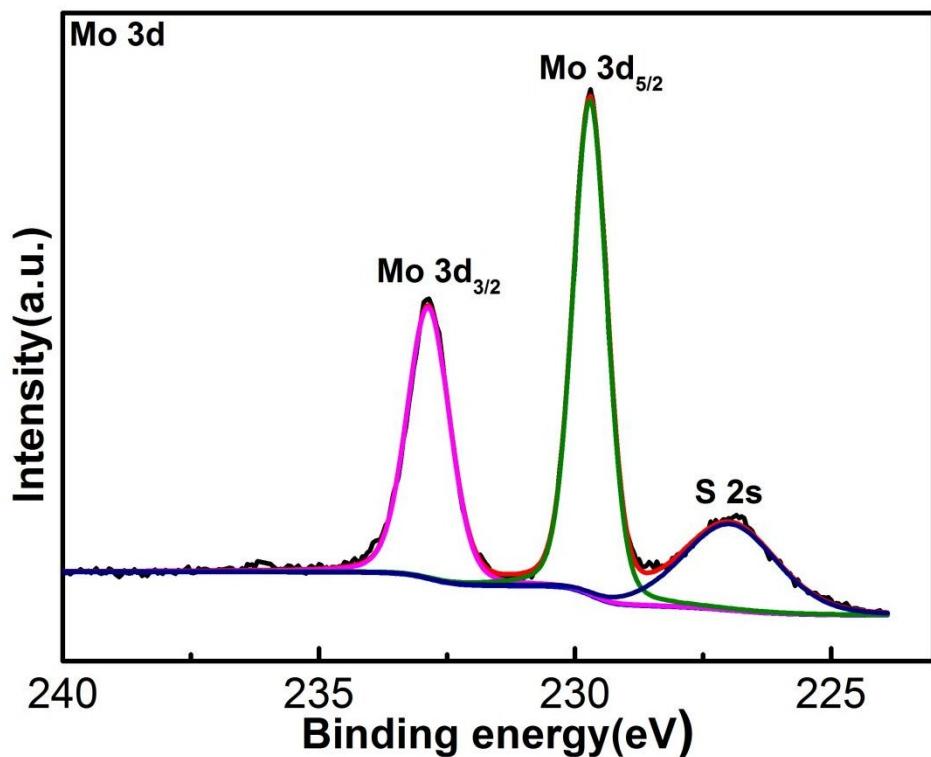
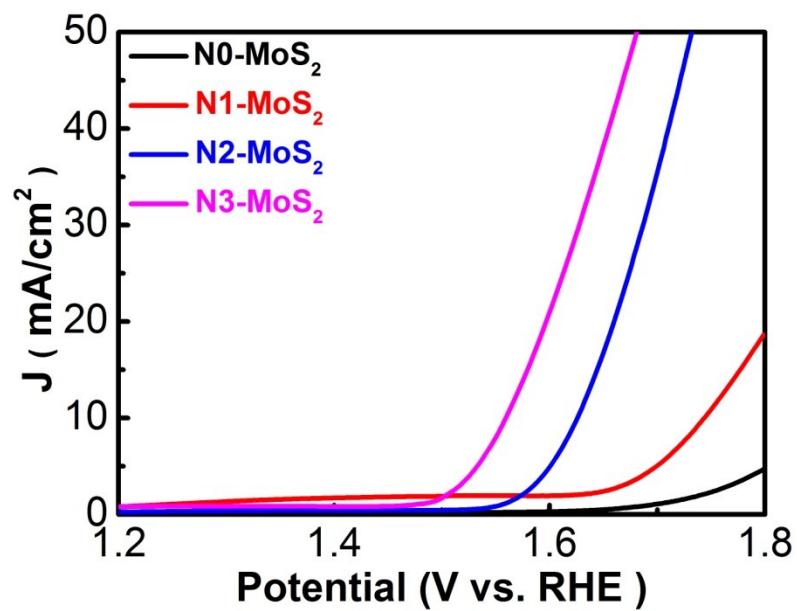


Fig. S5 The XPS spectra of Mo orbitals for N3-MoS<sub>2</sub>

**Table S1 The concentration of elements in four samples before ECT**

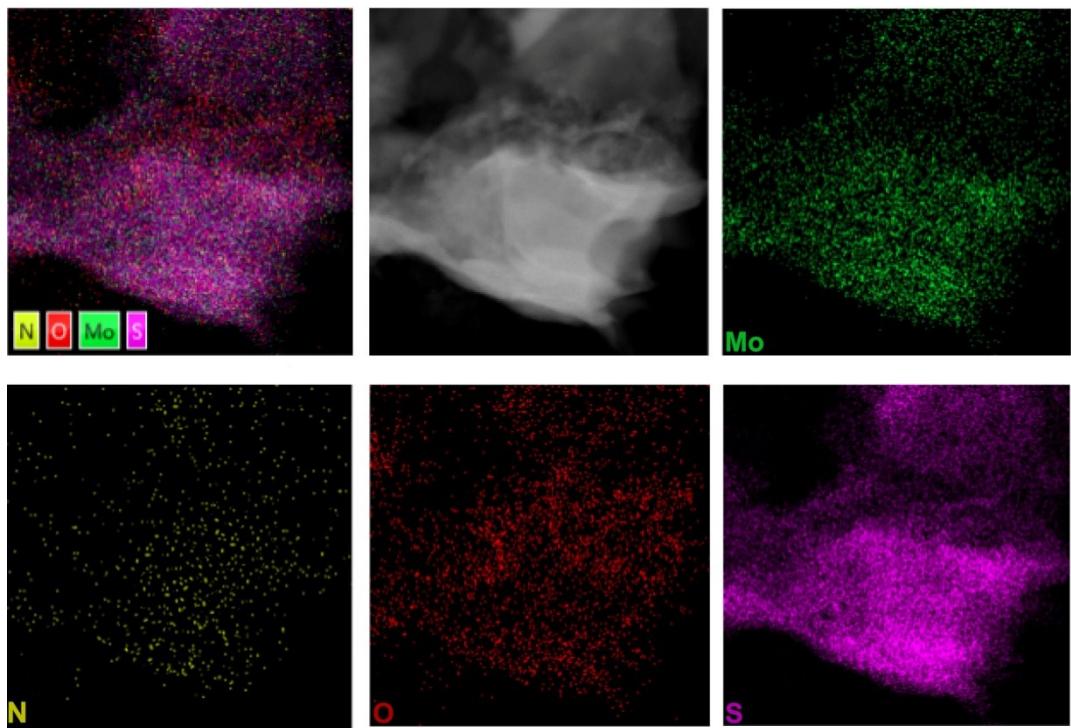
Catalyst	Mo (Atomic%)	S (Atomic%)	N (Atomic%)	O (Atomic%)
N0-MoS2	28.5	70.76	0.65	0.09
N1-MoS2	28.21	70.03	1.68	0.08
N2-MoS2	27.96	69.12	2.85	0.07
N3-MoS2	27.6	69.06	3.26	0.08



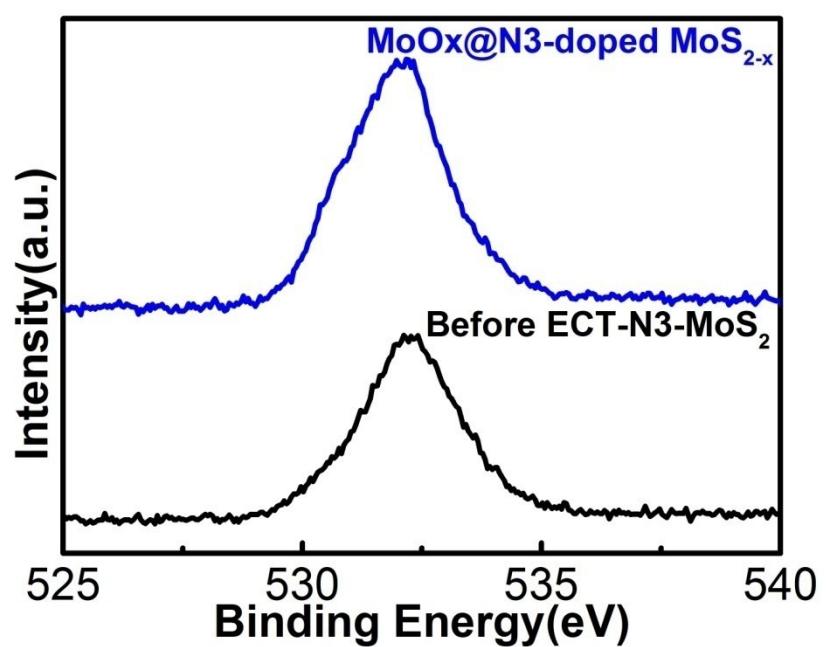
**Fig. S6** Polarization curves of N0-MoS<sub>2</sub>, N1-MoS<sub>2</sub>, N3-MoS<sub>2</sub> and N4-MoS<sub>2</sub>

**Table S2 The concentration of elements in MoOx@N3-doped MoS<sub>2-x</sub> and Before-ECT-MoS<sub>2</sub>**

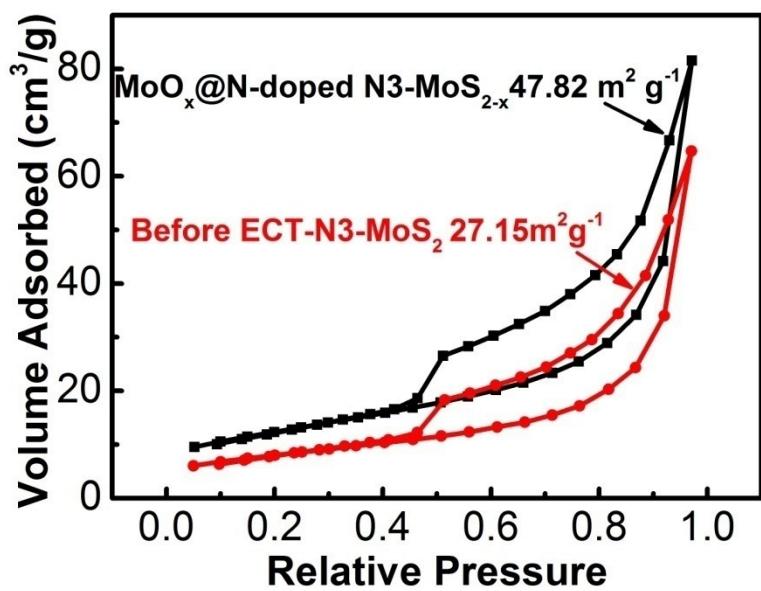
Catalyst	S (Atomic%)	N (Atomic%)	O (Atomic%)
	Mo (Atomic%)		
Before-ECT-MoS <sub>2</sub>	27.6	69.06	3.26
MoOx@N3-doped MoS <sub>2-x</sub>	26.86	68.18	3.12



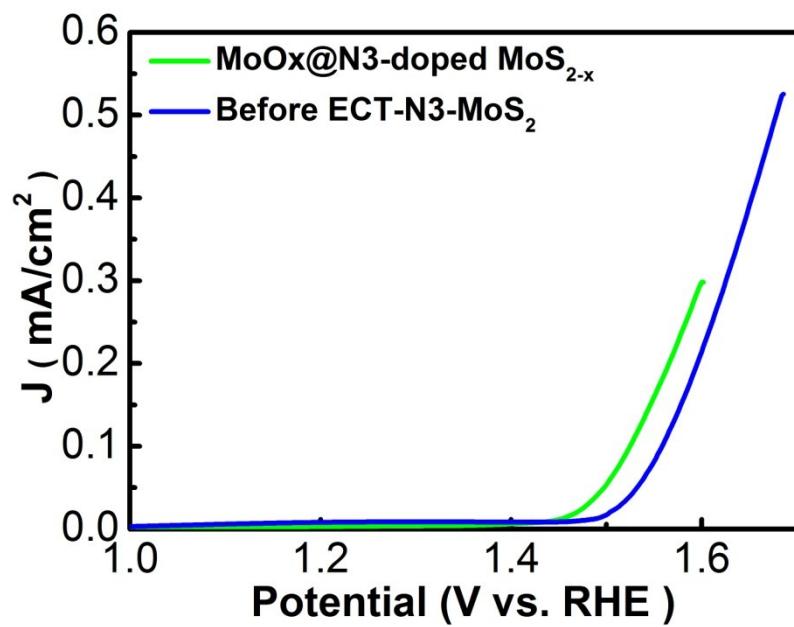
**Fig. S7** The corresponding energy-dispersive X-ray spectroscopy (EDX) mapping MoO<sub>x</sub>@N<sub>3</sub>-doped MoS<sub>2-x</sub>.



**Fig. S8 The XPS spectra of O orbitals for Before ECT-N3-MoS<sub>2</sub> and MoOx@N3-doped MoS<sub>2-x</sub>**



**Fig. S9** N<sub>2</sub> sorption isotherms of Before ECT-N3-MoS<sub>2</sub> and MoO<sub>x</sub>@N3-doped MoS<sub>2-x</sub>



**Fig. S10** The polarization curves normalized by the BET surface area

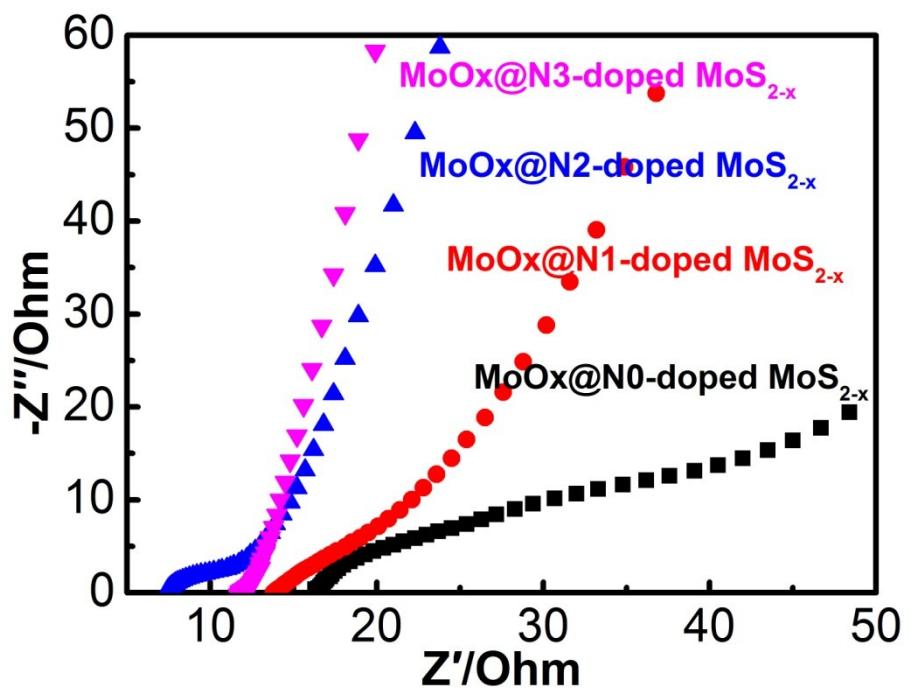
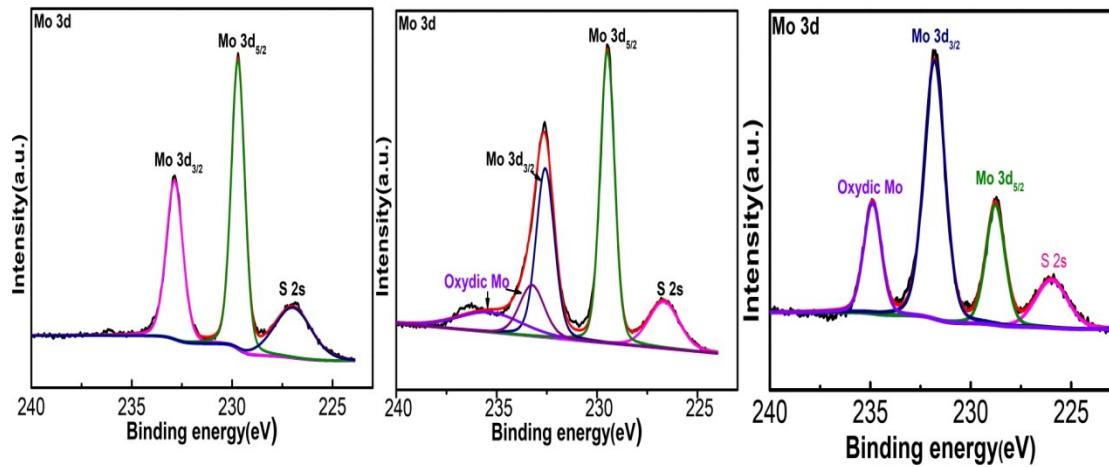


Fig. S11 Nyquist plots of the as-prepared MoS<sub>2</sub> nanosheets



**Fig. S12 The XPS spectra of Mo orbitals for Before ECT-N3-MoS<sub>2</sub>, MoOx@N3-doped MoS<sub>2-x</sub> and After durability test MoOx@N3-doped MoS<sub>2-x</sub>**

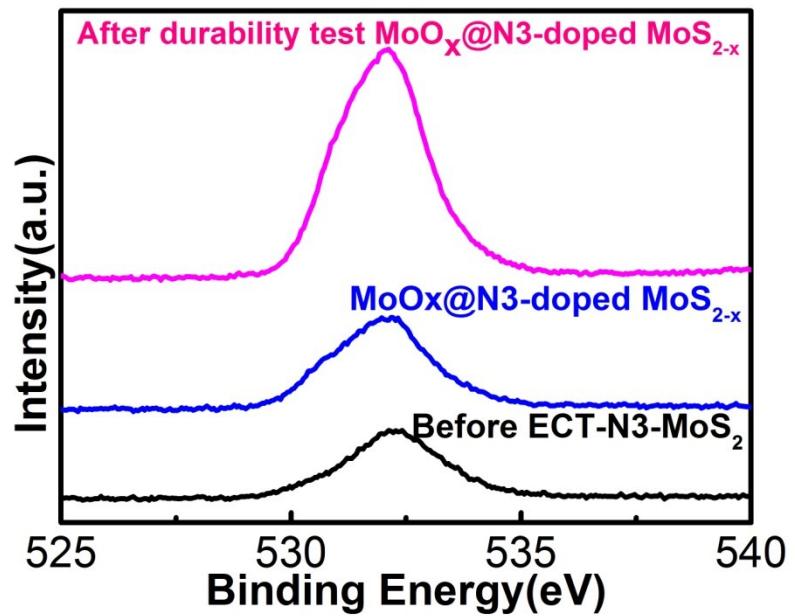
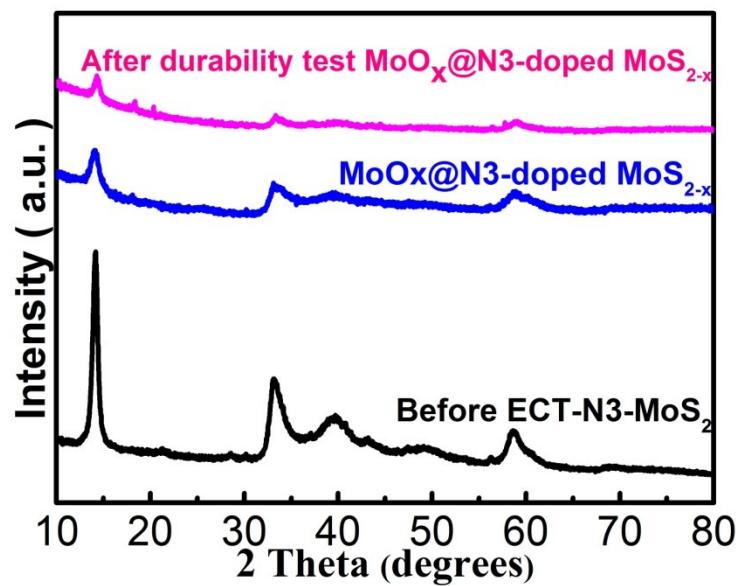
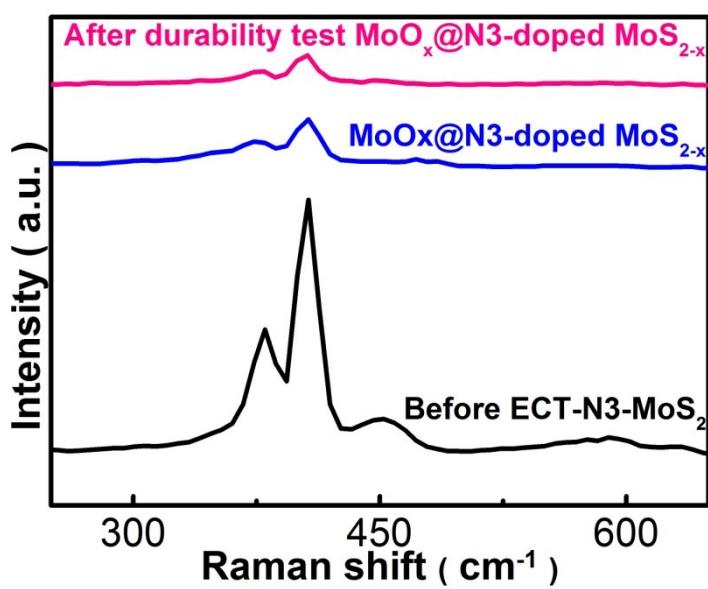


Fig. S13 The XPS spectra of O orbitals for Before ECT-N3-MoS<sub>2</sub>, MoO<sub>x</sub>@N3-doped MoS<sub>2-x</sub> and After durability test MoO<sub>x</sub>@N3-doped MoS<sub>2-x</sub>



**Fig. S14 XRD patterns of Before ECT-N3-MoS<sub>2</sub> , MoO<sub>x</sub>@N3-doped MoS<sub>2-x</sub> and  
After durability test MoO<sub>x</sub>@N3-doped MoS<sub>2-x</sub>**



**Fig. S15** Raman spectra of Before ECT-N3-MoS<sub>2</sub> , MoO<sub>x</sub>@N3-doped MoS<sub>2-x</sub> and After durability test MoO<sub>x</sub>@N3-doped MoS<sub>2-x</sub>

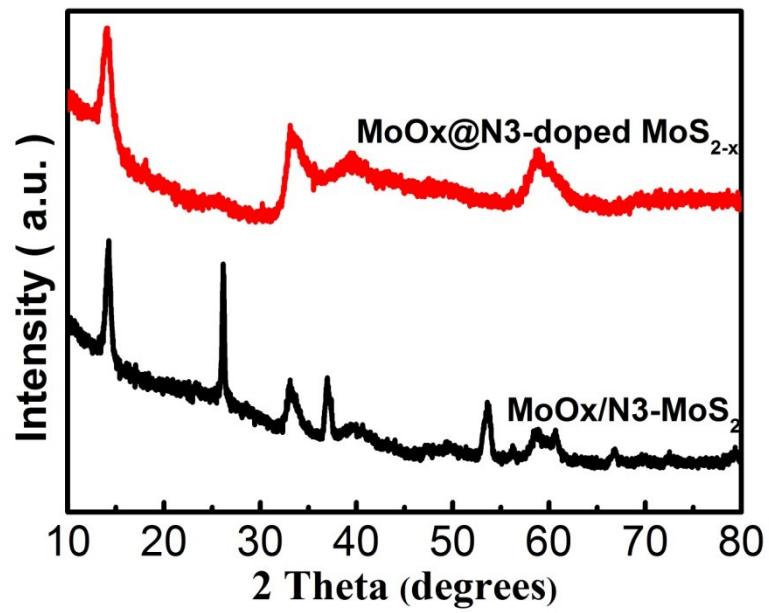
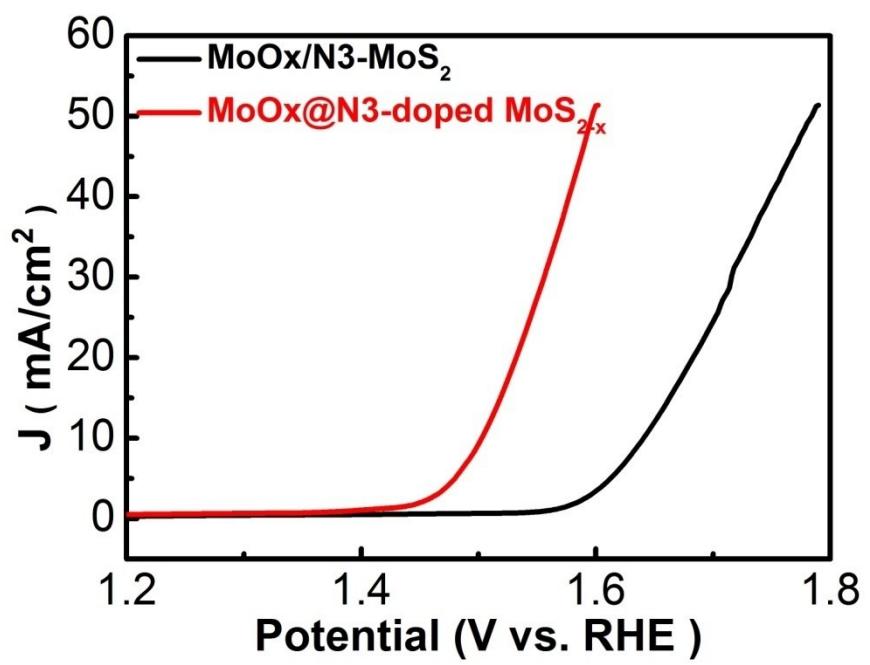
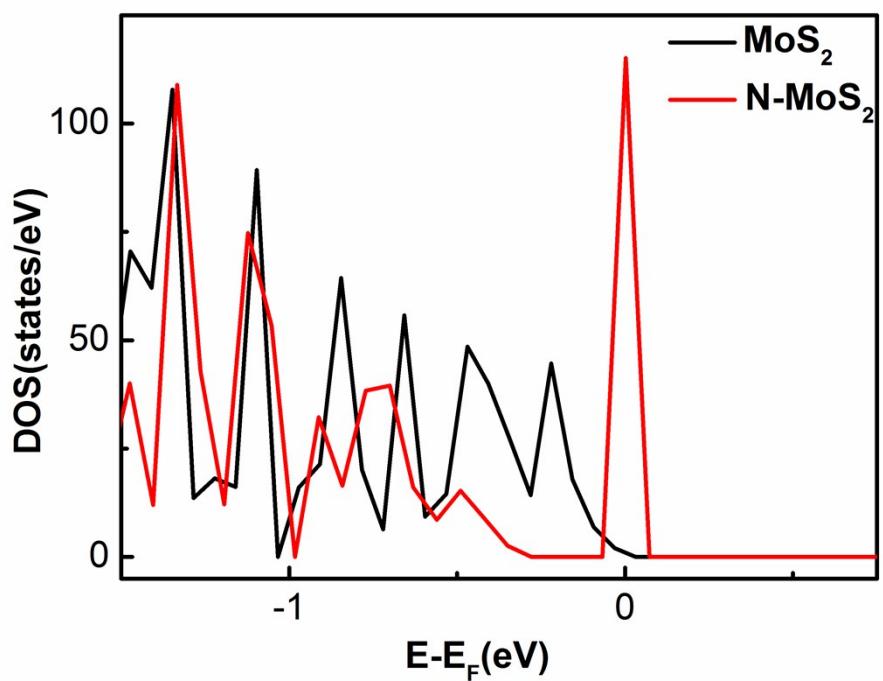


Fig. S16 XRD patterns of MoOx/N3-MoS<sub>2</sub> and MoOx@N3-doped MoS<sub>2-x</sub>



**Fig. S17** Polarization curves of MoOx/N3-MoS<sub>2</sub> and MoOx@N3-doped MoS<sub>2-x</sub> nanosheets



**Fig.S18 Density of states (DOS) plots of as-prepared  $\text{MoS}_2$  nanosheets.**

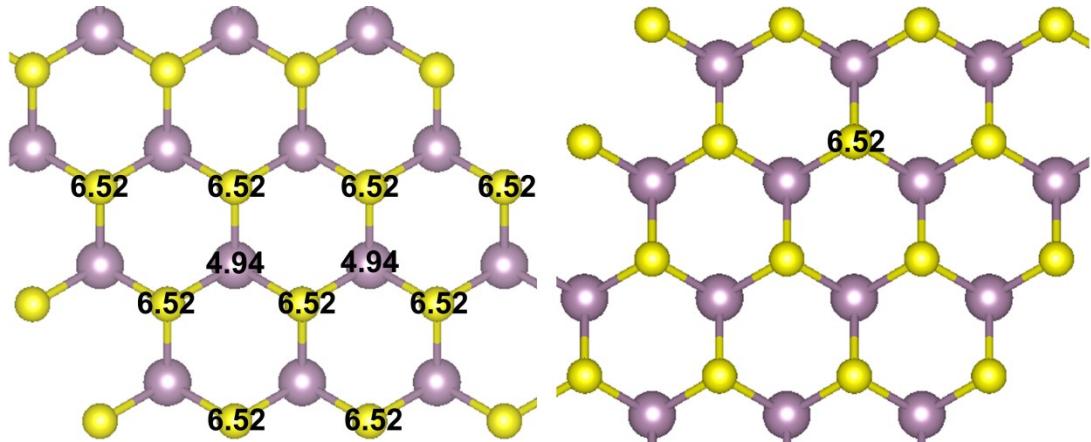


Fig. S19 Bader charge of different atoms for pristine MoS<sub>2</sub>

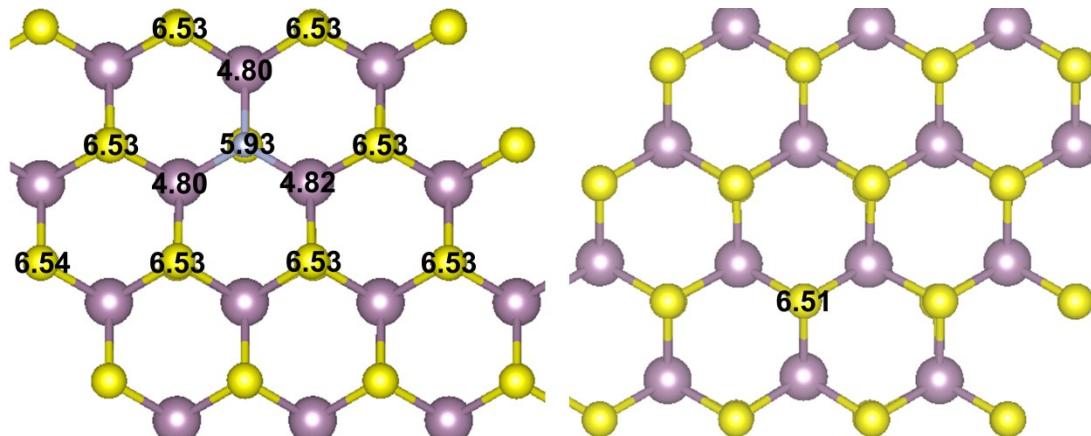
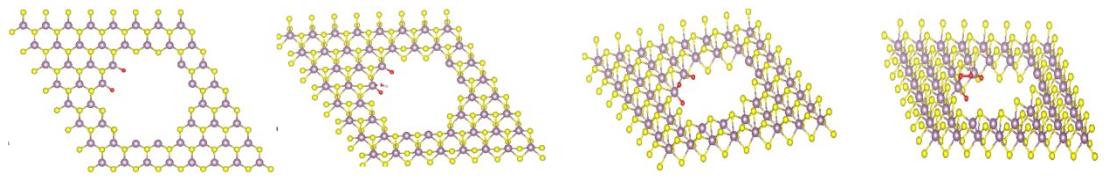
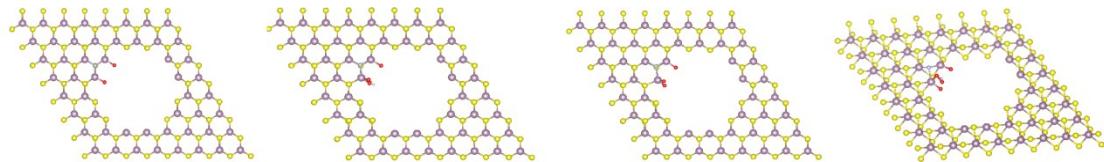


Fig. S20 Bader charge of different atoms for N-MoS<sub>2</sub>



**Fig.S21** the corresponding atomic configurations of the intermediate adsorption for pristine MoS<sub>2</sub>



**Fig.S22** the corresponding atomic configurations of the intermediate adsorption for N-MoS<sub>2</sub>

**Table S3. Comparison of the OER activity for several recently reported highly active metal-free catalysts supported on different substrates.**

Catalyst	Potential @ 10.0mA cm-1 (Vs. RHE)	Tafel slope (mV dec <sup>-1</sup> )	Electrolyte	Substrate	Reference
Benchmarking	1.53	70	1 M KOH	Glassy carbon	This work
RuO <sub>2</sub>					
N3-MoS <sub>2</sub> -ECT	1.50	61	1 M KOH	Glassy carbon	This work
single-unit-cell	~1.6	64	1 M KOH	Glassy carbon	[1]
thick CoSe <sub>2</sub> sheets					
NCNT/CoxMn <sub>1-x</sub> O	1.57	40	1 M KOH	Glassy carbon	[2]
Co <sub>3</sub> O <sub>4</sub> /N-rmGO	1.54	67	1 M KOH	Glassy carbon	[3]
A-CoS <sub>4.6</sub> O <sub>0.6</sub>	1.52	62	1 M KOH	Glassy carbon	[4]
PNCs					
N-CoFe LDHs	1.511	40.03	1 M KOH	Glassy carbon	[5]
Co@CoOx	1.519	68.9	1 M KOH	Glassy carbon	[6]
N-CoS <sub>2</sub>	1.47	98	1 M KOH	Glassy carbon	[7]
N-NiS <sub>2</sub>	1.5	none	1 M KOH	Glassy carbon	[8]
FeV	1.48	36.7	1 M KOH	Glassy carbon	[9]
FeCoMo	1.507	27.74	1 M KOH	Glassy carbon	[10]

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