Supporting Information

Hierarchical 2D Ni-Mo-S nanosheets@nitrogen doped graphene hybrid as Pt-free cathode for high-performance dye sensitized solar cells and fuel cells

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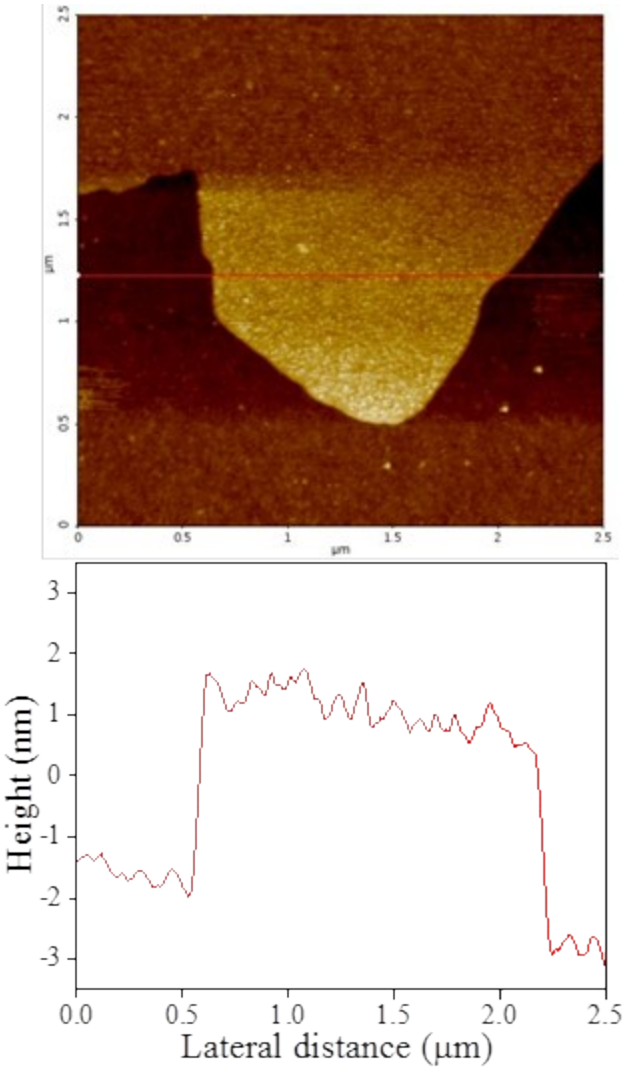


Fig. S1 AFM image and its height profile of Ni-Mo-S/NG hybrid indicating thickness of few nm and width ~2.5 μm.

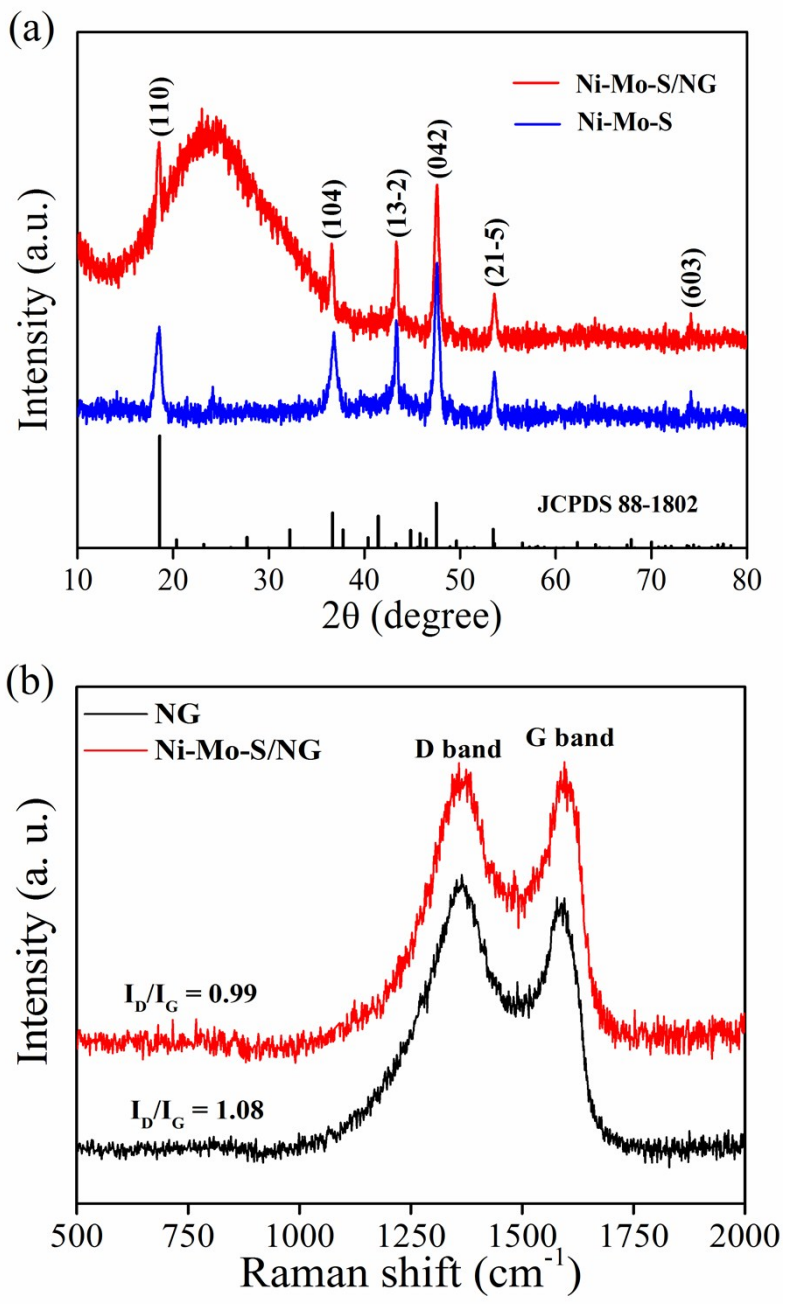
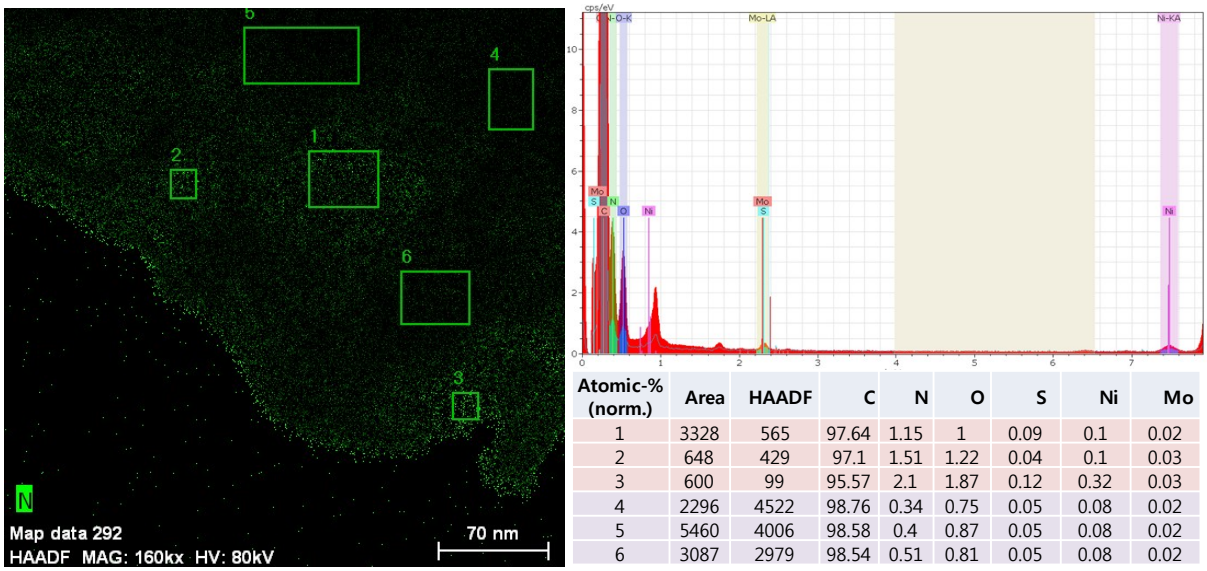


Fig. S2 (a) XRD pattern of as-synthesized Ni-Mo-S nanosheet and Ni-Mo-S/NG hybrid, and (b) Raman spectra of as-synthesized NG and Ni-Mo-S/NG hybrid.



Atomic-% (norm.)	Area	HAADF	C	N	O	S	Ni	Mo
1	3328	565	97.64	1.15	1	0.09	0.1	0.02
2	648	429	97.1	1.51	1.22	0.04	0.1	0.03
3	600	99	95.57	2.1	1.87	0.12	0.32	0.03
4	2296	4522	98.76	0.34	0.75	0.05	0.08	0.02
5	5460	4006	98.58	0.4	0.87	0.05	0.08	0.02
6	3087	2979	98.54	0.51	0.81	0.05	0.08	0.02

Fig. S3 EDS quantification MAP data (K-factor) and its corresponding atomic wt. % of the Ni-Mo-S/NG hybrid.

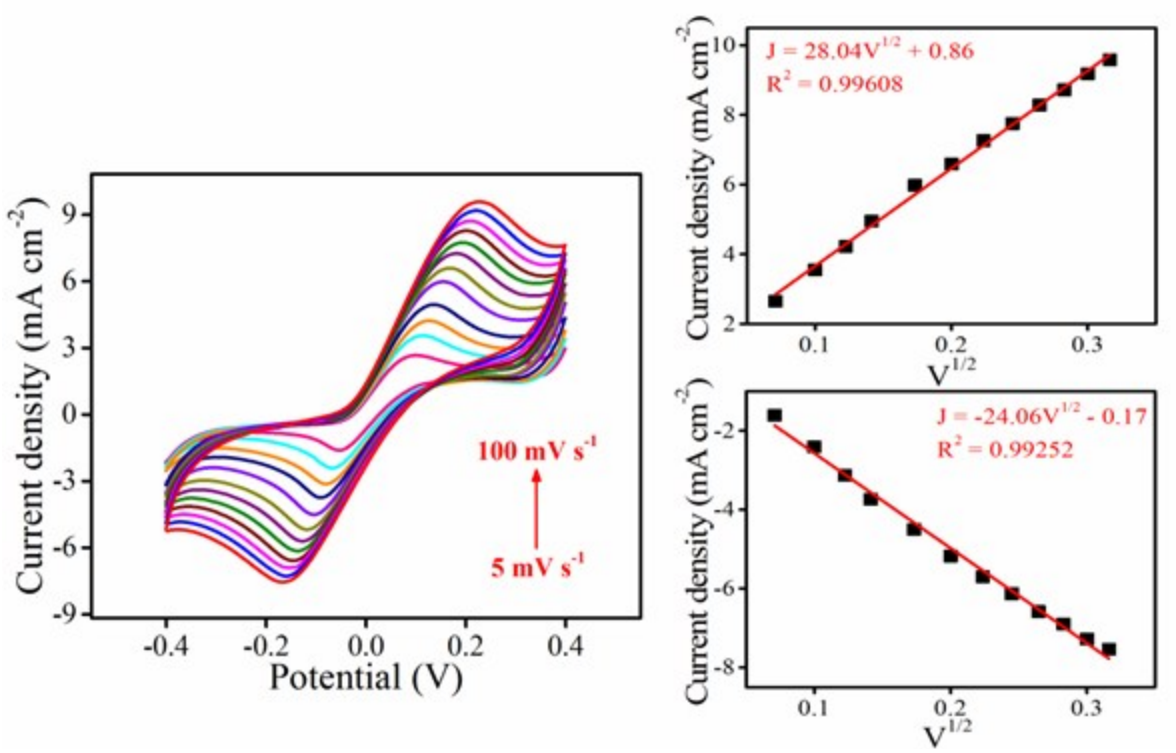


Fig. S4 (a) CV curves of the Ni-Mo-S/NG CE with different sweep rates, and b) the relationship of square root of scan rate *vs* redox peak current.

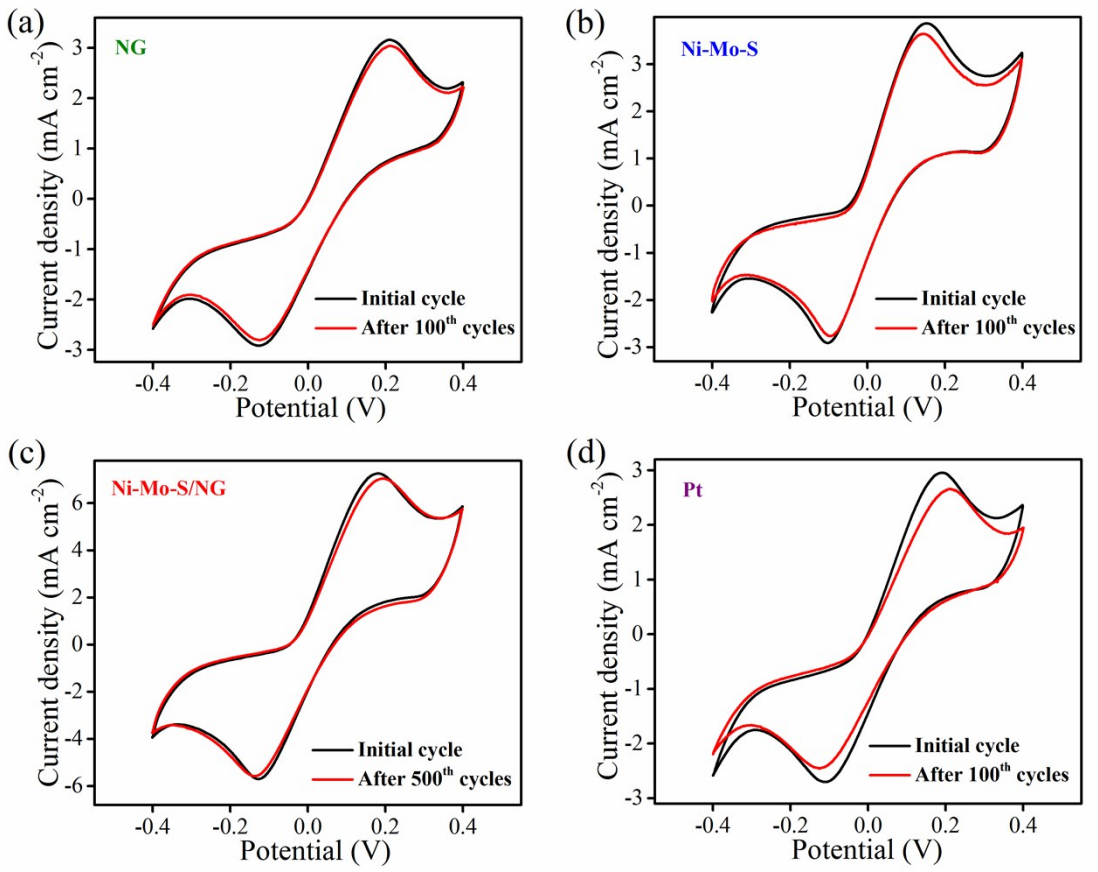


Fig. S5 Electrochemical stability of the NG, pure Ni-Mo-S, Ni-Mo-S/NG and Pt was investigated by successive CV scanning towards I^-/I_3^- reduction reaction (scan rate: 50 mV s^{-1}).

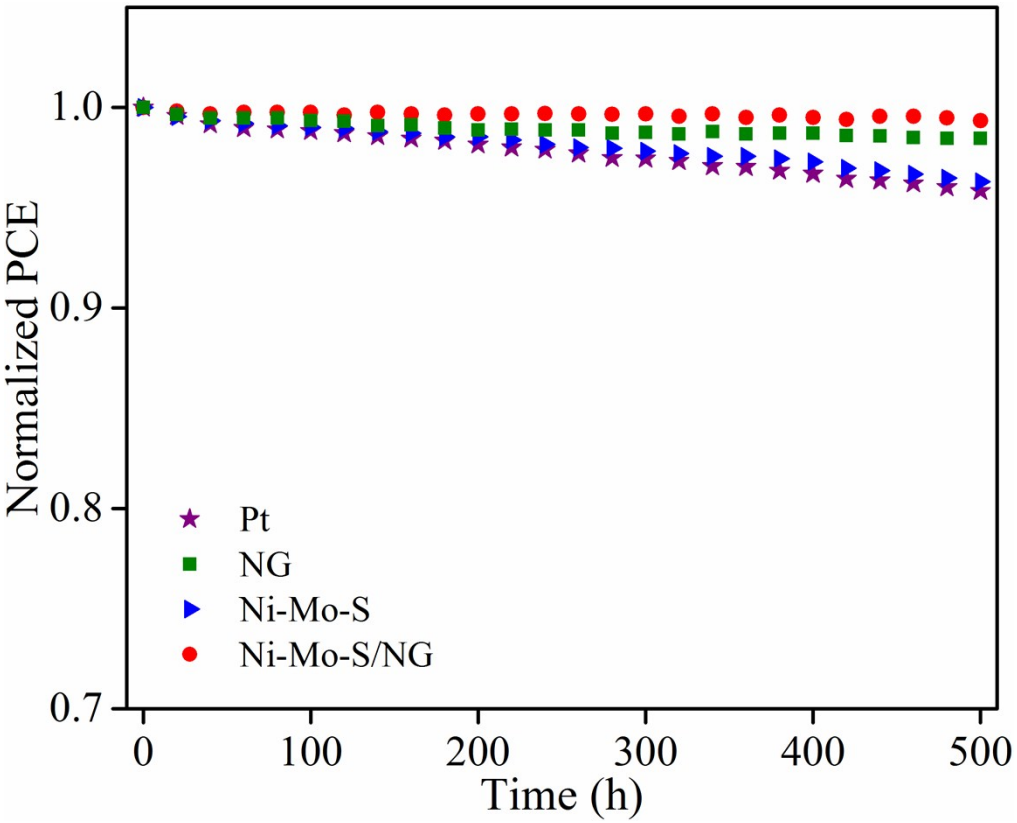


Fig. S6 Long-term stability test for DSSCs based on NG, pure Ni-Mo-S, Ni-Mo-S/NG and Pt CEs.

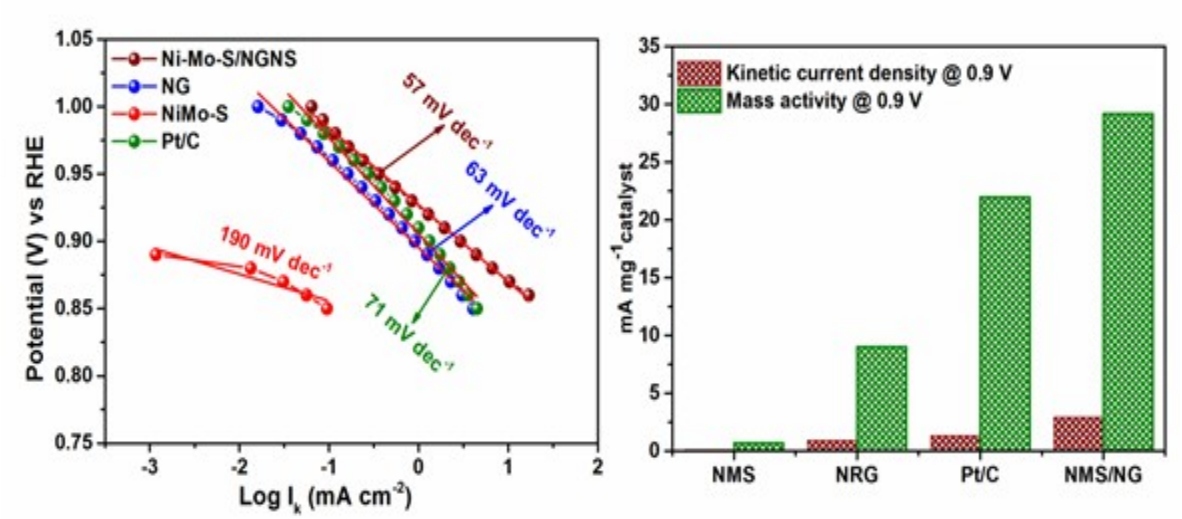


Fig. S7 (a) tafel plots (b) kinetic current density and mass activity of Ni-Mo-S, NG, Pt/C and Ni-Mo-S/NG catalysts measured at a mass transport corrected potential of 0.9 V vs RHE. (c) Initial and 2000th CVs Pt/C catalyst recorded in N_2 saturated 0.1 M KOH at a scan rate of 50 mV S^{-1} (d) LSVs of Pt/C catalyst recorded in O_2 saturated 0.1 M KOH at a scan rate of 10 mV S^{-1} with an electrode rotation speed of 1600 rpm.

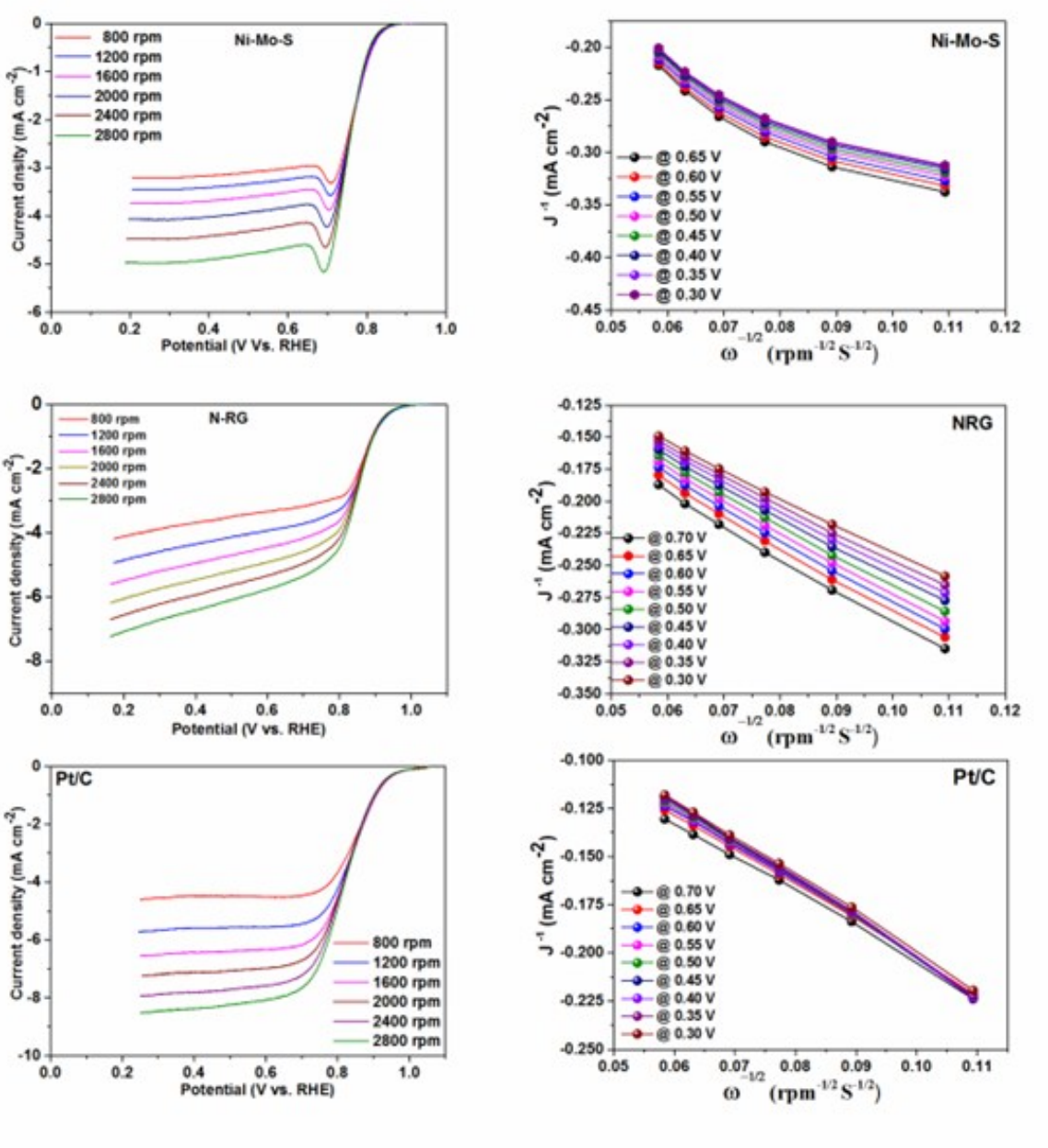


Fig. S8 LSV recorded at different rotation speeds and corresponding K-L plots of the catalyst (a), (c), (e), and (b), (d) and (f) of Ni-Mo-S, NG and Pt/C catalysts.

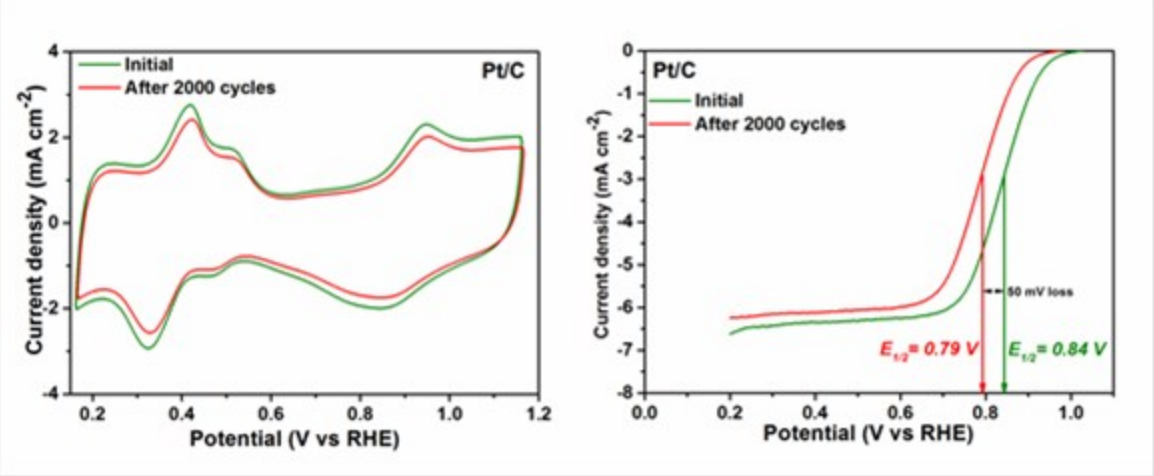


Fig. S9 (a) Initial and 2000th CVs Pt/C catalyst recorded in N_2 saturated 0.1 M KOH at a scan rate of 50 mV S^{-1} (b) LSVs of Pt/C catalyst recorded in O_2 saturated 0.1 M KOH at a scan rate of 10 mV S^{-1} with the electrode rotation speed of 1600 rpm.

Table S1. Elemental composition of as-synthesized NG, pure Ni-Mo-S, and Ni-Mo-S/NG estimated from ICP-AES measurements.

Sample	Ni (at. %)	Mo (at. %)	S (at. %)	N (at. %)	O (at. %)	C (at. %)
NG	-	-	-	9.87	7.15	82.98
Ni-Mo-S	28.89	27.95	32.45	-	10.71	-
Ni-Mo-S/NG	3.62	3.54	9.06	10.24	6.29	67.25

Table S2. Photovoltaic parameters of DSSCs with different CEs under 100 mW cm^{-2} illuminations.

Counter electrode	J_{sc} (mA cm^{-2})	V_{oc} (V)	FF	η (%)	References
$CoNi_2S_4$–CF	15.30	0.68	0.67	7.03	1
FeS_2	15.14	0.71	0.68	7.31	2
Co_9S_8/FTO	14.21	0.71	0.69	7.00	3
$Ni_{0.85}Se$	16.67	0.74	0.64	7.85	4
CuInS–ZnS	15.3	0.77	0.64	7.50	5
$Co_{0.85}Se$	16.98	0.74	0.75	9.40	6
$Ni_{0.85}Se$	15.63	0.74	0.72	8.32	6
CuZnSnSSe	17.70	0.80	0.52	7.37	7
CoS	14.70	0.78	0.71	8.10	8
NiS_2@RGO	16.55	0.75	0.69	8.55	9
Cu_2FeSnS_4	14.57	0.75	0.73	8.03	10
NiS	14.70	0.73	0.66	6.81	11
NiS/graphene	10.31	0.72	0.70	5.25	12
Patterned MoS_2	15.2	0.73	0.52	5.80	13
MoS_2-C	15.07	0.75	0.68	7.69	14
NHGF	11.33	0.75	0.66	5.56	15
NGC	16.23	0.77	0.54	6.74	16
L-GQDs	15.21	0.76	0.72	7.70	17
MoS_2/RGO–CNT	14.59	0.76	0.67	7.46	18

MoS_2–GNS	12.79	0.77	0.59	5.81	19
MoS_2/RGO	12.51	0.73	0.66	6.04	20
G–MoS_2	16.1	0.66	0.67	7.10	21
N-GNRs	15.18	0.78	0.72	8.57	22
N-MEG	15.04	0.73	0.65	7.18	23
CNx/CNT	16.30	0.73	0.61	7.38	24
GNR/CNT	16.73	0.73	0.67	8.23	25
Pt	14.54	0.77	0.78	8.73	This work
Ni-Mo-S/NG	15.65	0.79	0.80	9.89	This work

NHGF – nitrogen doped holey reduced graphene; NGC – N-doped graphene-CNT; L-GQDs – laser-reduced graphene quantum dots; G-MoS_2 – Graphene-mediated highly-dispersed MoS_2; GNR – graphene nanoribbon; N-GNRs – nitrogen doped graphene nanoribbon; N-MEG – nitrogen-doped microwave-exfoliated graphene

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