Supporting information

Experimental section

Preparation of $(NH_4)_2MoS_4$

Ammonium tetrathiomolybdate ($(NH_4)_2MoS_4$) was obtained by ammonium molybdate ($(NH_4)_6Mo_7O_{24}\cdot 4H_2O$) and ammonium sulfide ($(NH_4)_2S$) according to a previously reported method. In a standard experimental procedure, 10 g $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ (0.008 mol) was dissolved in 40 mL distilled(DI) water and subsequently combined with 12 mL of ammonia solution. The resulting mixture was heated to 45 ℃ and stirred for 30 minutes. Then, 108 mL $(NH_4)_2S$ solution of 14% mass fraction was added to the above solution, stirred at 65 ℃ for 1 h to obtain blood red liquid. The reaction was cooled to 0 °C for crystallization for 6 h, and then filtered with a Brinell funnel. During the filtering, 240 mL cold DI water was used for washing three times, and then 120 mL anhydrous ethanol was used for washing three times. The product was dried in a vacuum drying oven at 50 °C for 12 h. The product was a brownish red needle-like solid, which was $(NH_4)_2MoS_4$. The reaction equation in the synthesis process is as follows:

$$(NH_4)_6Mo_7O_{24}\cdot 4H_2O + 8NH_3\cdot H_2O \rightarrow 7(NH_4)_2MoO_4 + 8H_2O \quad (1)$$

$$(NH_4)_2MoO_4 + 4(NH_3)_2S \rightarrow (NH_4)_2MoS_4 + 4NH_3\cdot H_2O \quad (2)$$

Preparation of the MoS_2 films

MoS_2 was prepared by electro-deposition method on ITO-PET flexible substrate. The electrolyte consists of 5 mM $(NH_4)_2MoS_4$ and 0.1M KCl in DI water. Graphite plate and ITO-PET substrate were used as the anodic and cathodic electrodes. The electro-deposition voltage was controlled at 1.1V. MoS_2 was prepared by electro-deposition with a series times (MoS_2-x min, x:2, 5, 10, 20) at different temperatures (MoS_2-x ℃, x: 15, 20, 25). For the time-dependent experiments, the temperature was kept at 20 ℃. And the temperatures-dependent experiments, the time was kept at 10 min, while the electro-deposition temperature was changed as desired. The Pt counter electrode was prepared by electrodeposition as a reference.

Synthesis of etching MoS_2 films

MoS_2/ITO-PET was immersed in H_2O_2 solution with a series of temperatures (MoS_2-x ℃, x: 0, 15, 25, 35) inside which MoS_2/ITO-PET was allowed to react with H_2O_2 for varied concentrations (MoS_2-x mol/L, x:0.01, 0.1,1) at different times (MoS_2-x s, x:30, 60, 90, 120). For the temperature-dependent experiments, the time and solution concentration were kept at 60 s and 0.1 mol/L, while the H_2O_2 temperature was changed as desired. In the same way, for the concentration dependent experiments, the time and temperature were kept at 60 s and 15 ℃, while the H_2O_2 concentration was changed as at the set. For the time-dependent experiments, the temperature and solution concentration were kept at 15 ℃ and 0.1 mol/L, while the H_2O_2 time was changed as needed.

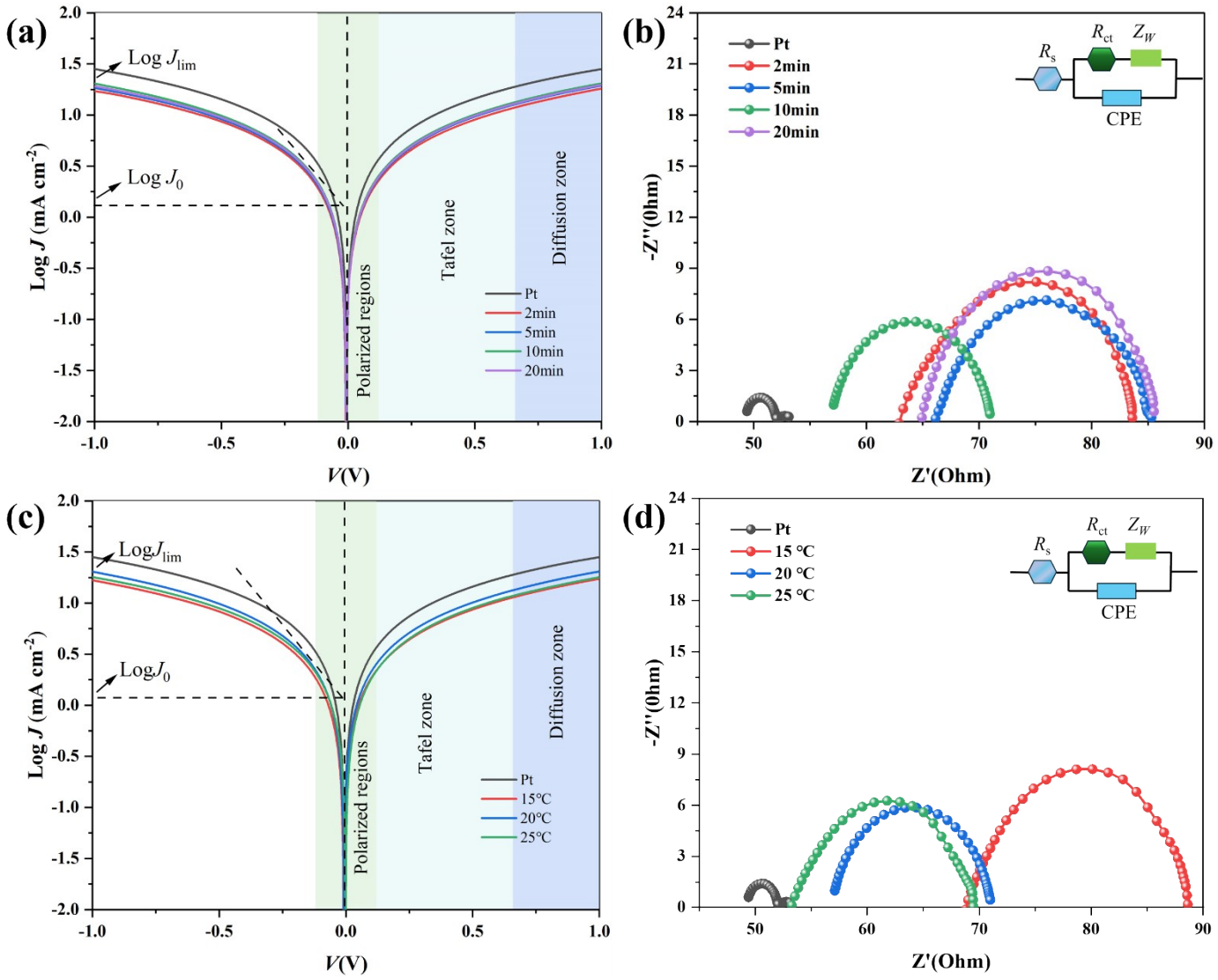
Characterization

The morphologies of MoS_2 nanoparticle were characterized by SEM (Hitachi S-8010, Japan) at an accelerating voltage of 5kV. The detailed information of MoS_2 element compositions were obtained by EDX. XRD (D/teX UItra 250) measurements were performed to the crystal structure of MoS_2 film in the range of 10-80° and the test speed was 5°/min. Raman (HORIBA EVOLUTION) spectroscopy measurements are made at excitation wavelength of 532 nm and test range of 100-600cm^{-1}. The chemical states of molybdenum and sulfur were measured by XPS (ECASA LAB 250Xi) photoelectron spectrometer. To explore the effect of etching treatment on MoS_2 transparency, the UV-vis (jinghua UV-1800PC) adsorption spectra were measured. The FTIR (IRAffinity-1S) spectra of MoS_2 before and after etching were measured.

Electrochemical Measurements and Cell fabrication

The Tafel curve, Electrochemical impedance spectra (EIS) and current density-time (I-t) were measured by an electrochemical workstation (CHI 660E, Shanghai Chenhua). Cyclic voltammograms were obtained in acetonitrile solution containing 10 mm LiI, 1 mm I_2, and 0.1 m $LiClO_4$ at 50 mV s^{-1} with Ag/Ag^+ electrode, Pt electrode and prepared electrode were used as reference electrode, counter electrode and working electrode.

After the sintering of the prepared photoanode, $TiCl_4$ treatment was performed at 450 ℃ for 30 min. When it cools down, the $TiCl_4$-treated photoanode was dipped in 0.3 mM N719 dye as explained above for 24 h. The CEs were prepared by electrodeposition on ITO-PET substrates. The electrolyte solution containing 0.03 M I_2, 0.05 M LiI, 0.1 M guanidinium thiocyanate, 0.5 M 4-tert-Butylpyridine and 0.6 M 1,3-dimethylimidazoullum iodide in acetonitrile and pentonitrile. The photoelectrode and counter electrodes were assembled together by placing a thermal adhesive film (25 μm Surlyn, Solaronix) in between the electrodes. An electrolyte is then added between the photoanode and counter electrode to assemble the DSSCs. Photocurrent-voltage (*J-V*) curves of DSSCs were measured under simulated solar illumination (AM 1.5, 100 mW cm^{-2}). In order to investigate the application of DSSCs for indoor photovoltaic applications, the photocurrent-voltage (*J-V*) curves of DSSCs were measured under simulated indoor light (1000 lux).



57

58 Figure S1. a-b) Tafel and EIS plots of electrodeposited MoS_2 at different times; c-d) Tafel and EIS plots of electrodeposited

59 MoS_2 at different temperatures.

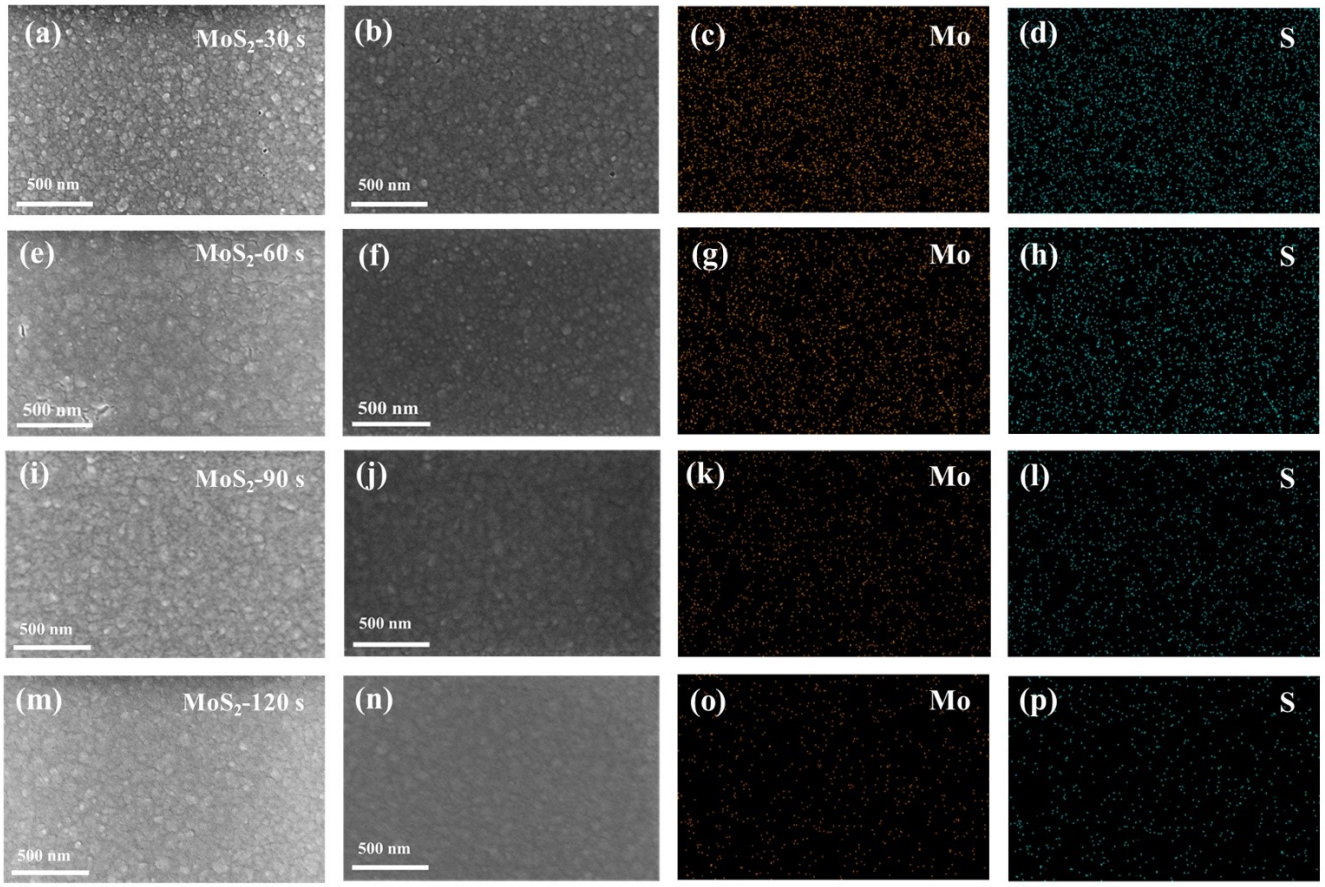


Figure S2. a) SEM images of MoS_2-30 s films; b-d) elemental mapping of MoS_2-30 s films; e) SEM images of MoS_2-60 s films; f-h) elemental mapping of MoS_2-60 s films; i) SEM images of MoS_2-90 s films; j-l) elemental mapping of MoS_2-90 s films, m) SEM images of MoS_2-120 s films; n-p) elemental mapping of MoS_2-120 s films.

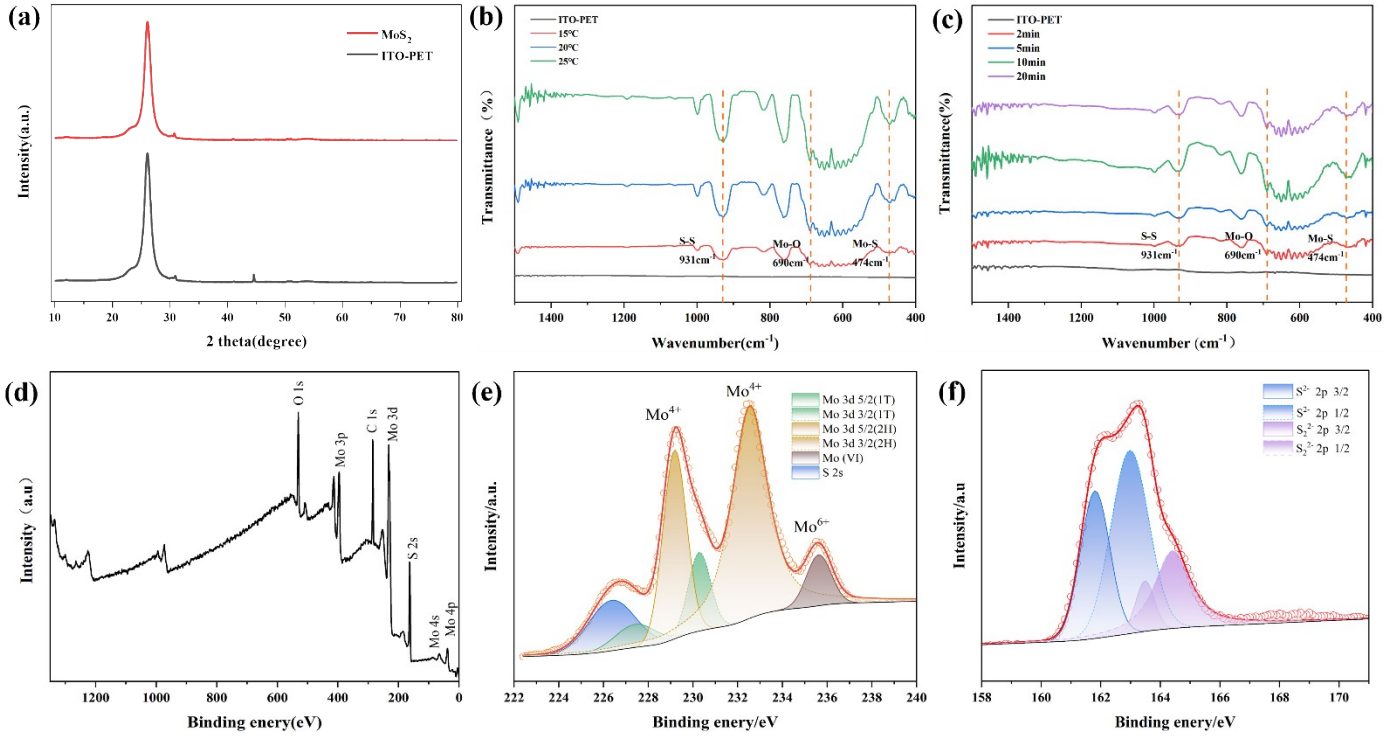


Figure S3. a) XRD patterns of ITO-PET and unetched MoS_2; b and c) FTIR spectra of MoS_2 by electrodeposition method for different temperatures and times; d) XPS spectra of unetched MoS_2; e,f) Mo 3d spectra and S 2s spectra of unetched MoS_2.

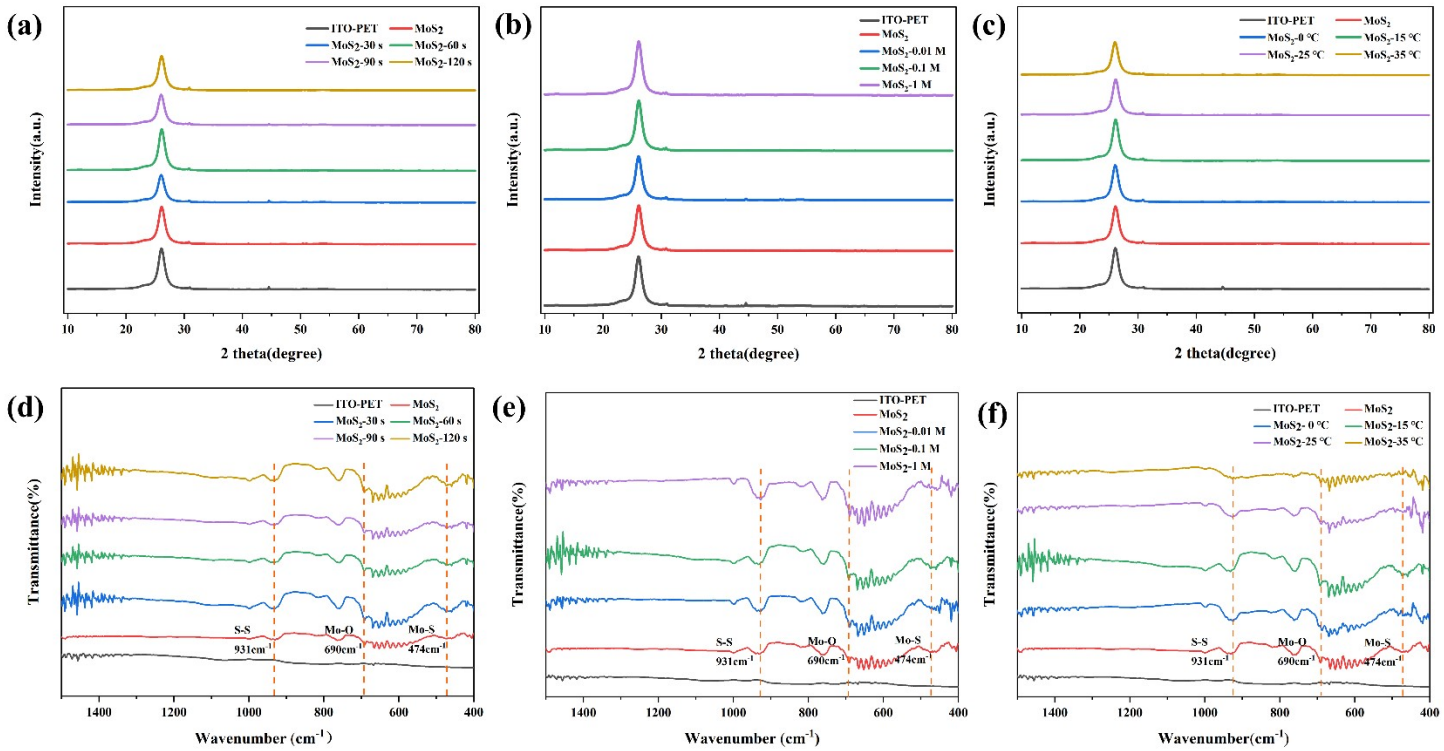
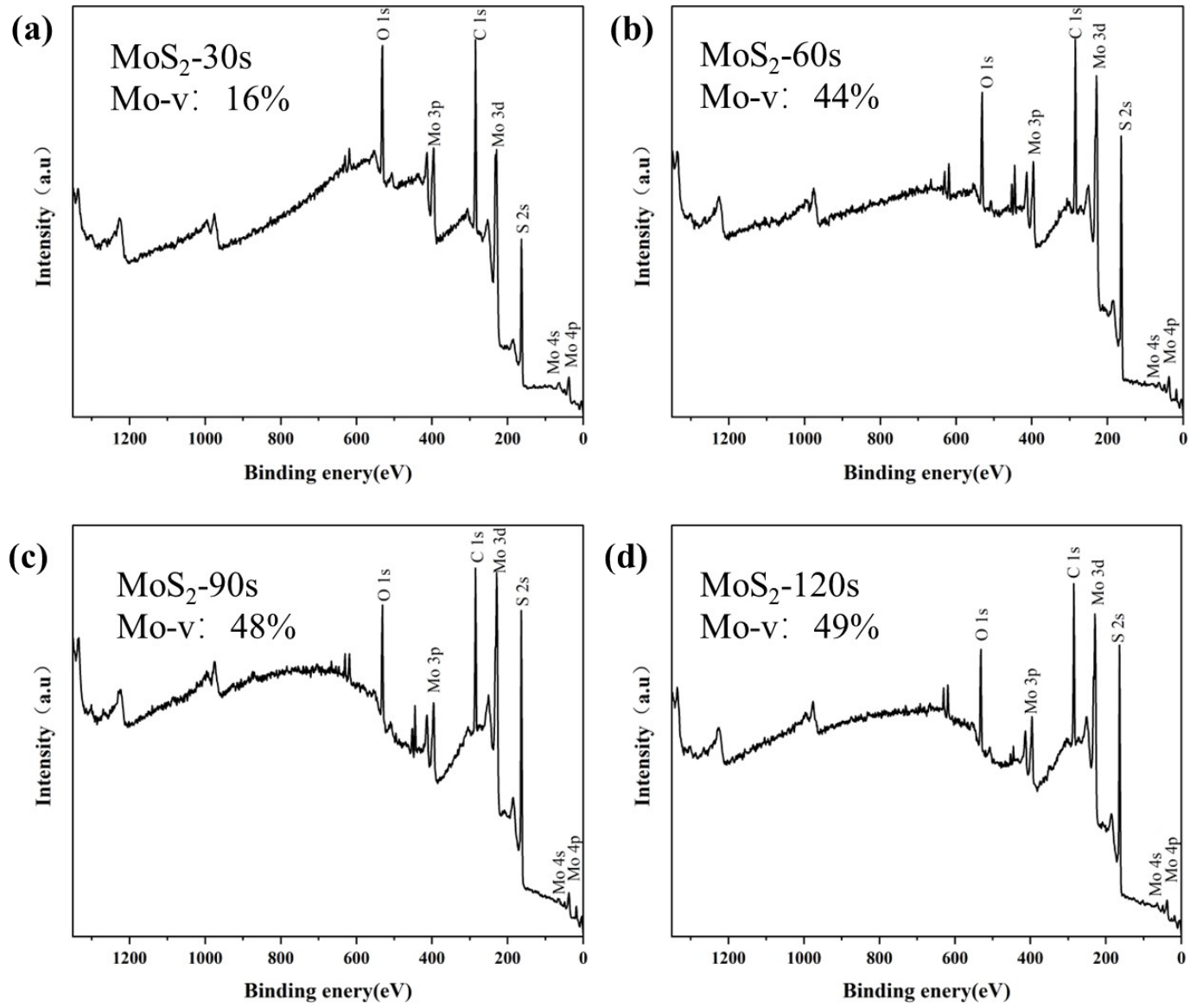
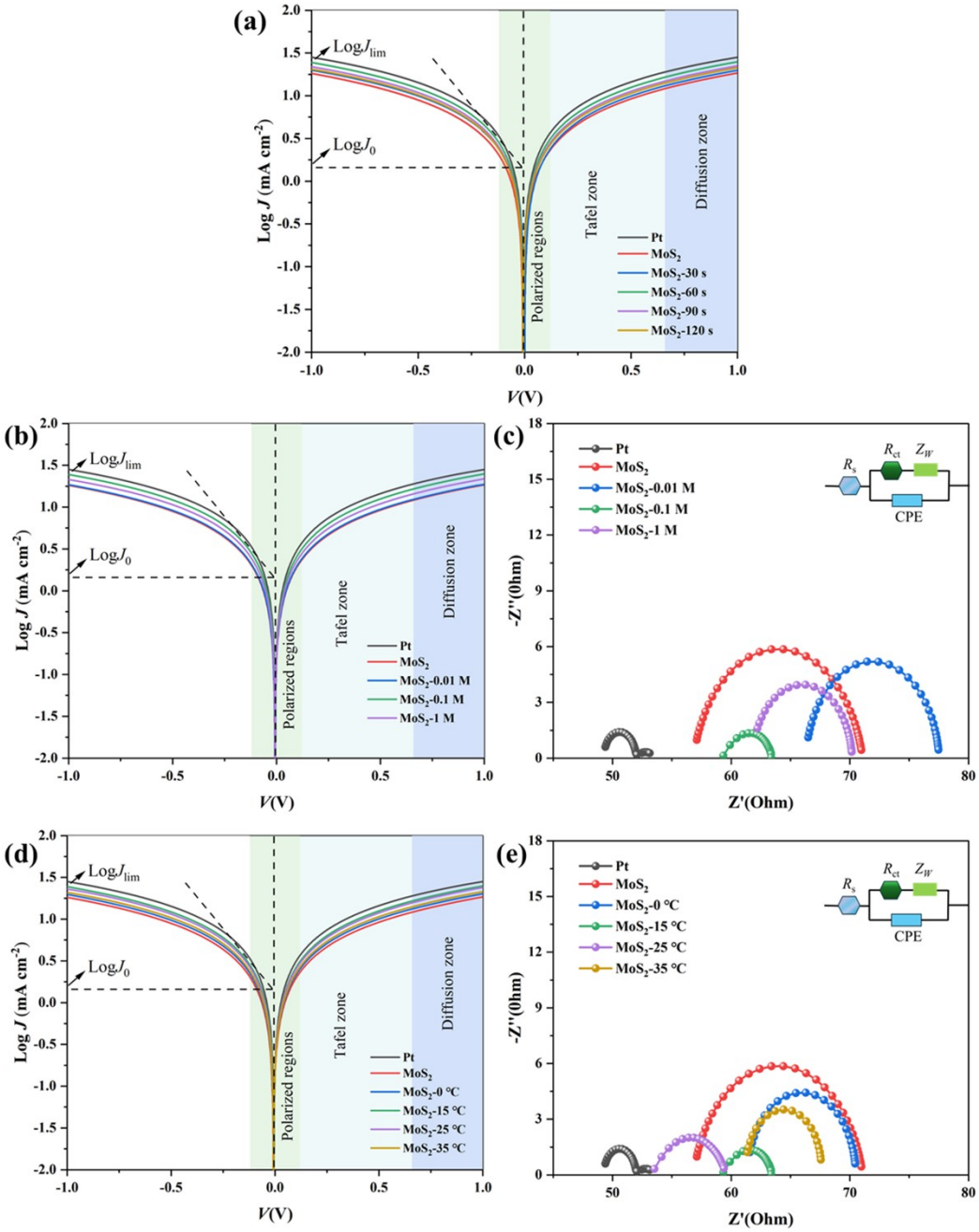


Figure S4. a, b, c) XRD patterns of unetched MoS_2 and etched MoS_2 by different etching times, etching concentrations and etching temperatures; d, e, f) FTIR spectra of unetched MoS_2 and etched MoS_2 by different etching times, etching concentrations and etching temperatures.



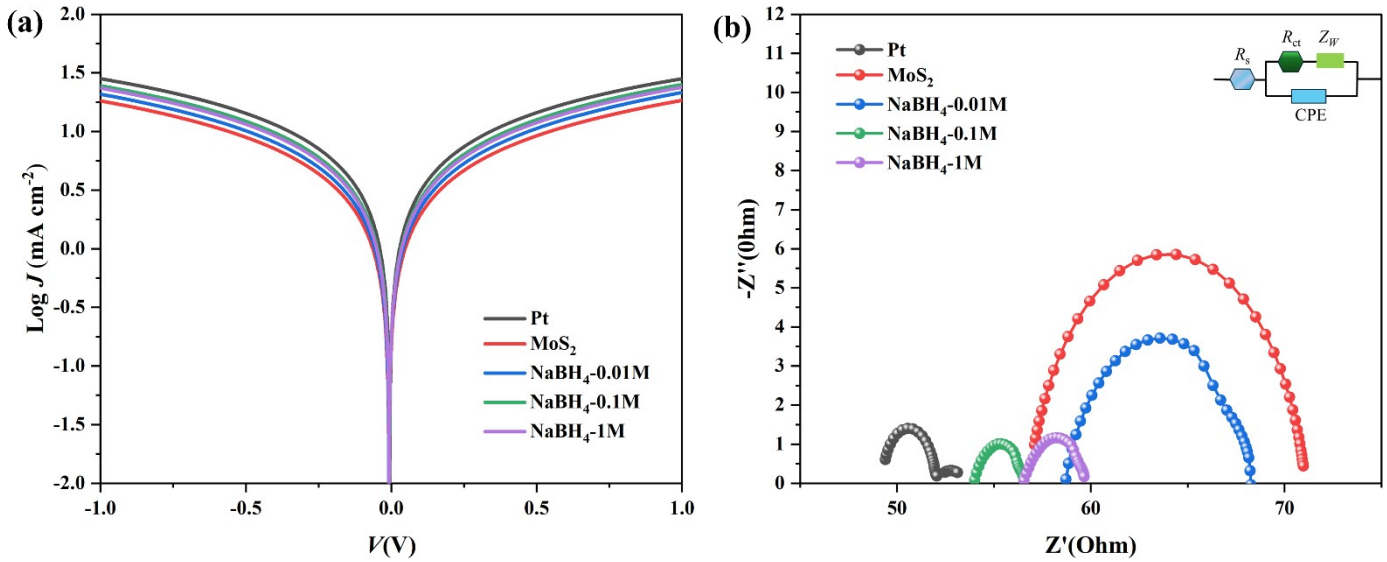
72

73 Figure S5. a, b, c, and d) XPS spectra of MoS_2-30 s, MoS_2-60 s, MoS_2-90 s and MoS_2-120 s.



74

75 Figure S6. a) Tafel plots of etching MoS_2 at different times, b-c) Tafel and EIS plots of etching MoS_2 at different
76 concentrations, d-e) Tafel and EIS plots of etching MoS_2 at different temperatures.



77

78 Figure S7. a,b) Tafel and EIS plots of MoS_2 etched with different concentrations of $NaBH_4$.