Supporting information

Layer-by-layer Self-Assembly of Polyelectrolyte Functionalized MoS₂ Nanosheets

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Figure S1. Histogram and typical AFM images of exfoliated MoS_2 nanosheets. The red line is fitted by Lorentz function.



Figure S2. SEM image of PAA-MoS $_2$ and PAM-MoS $_2$ nanosheets.



Figure S3. TEM images of liquid phase exfoliated MoS_2 with low (a) and high (b) resolution. Inset is the reduced FFT pattern of the corresponding HRTEM image.



Figure S4. Low resolution TEM images of typical PAA-MoS $_2$ (a) and PAM-MoS $_2$ (b) nanosheets.

From figure S4, note that the typical flakes of PAA-MoS₂ and PAM-MoS₂ are a few hundred nanometers in length (which is consistent with SEM images). In addition, the connection and partial coating of polymers on MoS_2 sheets can be clearly observed.



Figure S5. XPS survey spectra of PAA-MoS₂ (a) and PAM-MoS₂ (e). High-resolution XPS spectra of C 1s (b) and S 2p (c) of PAA-MoS₂; O1s spectrum of pure PAA (d); High-resolution XPS spectra of C 1s (f) and S 2p (g) of PAM-MoS₂; N1s spectrum of pure PAM (h)

XPS was used to determine the chemical components of the samples. Figure S5a shows the survey spectra of PAA-MoS₂. In the C 1s spectrum of PAA-MoS₂ (figure S5b), the binding energy of 284.5 eV is attributed to the C-C, C=C, and C-H bonds, while the deconvoluted peak center at 288.5 eV is a result of the O=C-N functional group. Furthermore, we ascribe the peak at 285.5 eV to the connection of PAA and MoS₂. As for the spectrum of S 2p (figure S5c), peaks at 163.2 and 162.0 eV can be ascribed to the S $2p_{1/2}$ and S $2p_{3/2}$ orbitals. These are consistent with the binding energies of Mo⁴⁺ and S²⁻ ions in the 2H MoS₂ phase. The O 1s spectrum of pure PAA (Figure S5d) exhibits a typical –COOH peak. Similar results are observable in the spectrum of PAM-MoS₂ (figure S5e-5g).



Figure S6. SEM images of the films after one (a), two (b), three (c), four (d), five (e) and six cycles (f).



Figure S7 TEM (a) and HRTEM (b) images of an LBL film after 3 absorption cycles.



Figure S8. UV-vis spectra of (PDDA/PAA-MoS₂/PAM-MoS₂/PEDT-PSS)_n LBL films (a) and (PDDA/ PEDT-PSS)_n LBL films assembled on glass slides.



Figure S9. TGA curves of the LBL films obtained with the pH values of PAA-MoS $_2$ and PAM-MoS $_2$ solutions as 10 and 2, respectively.



Figure S10. Cyclic voltammograms of bare ITO electrode and LBL films of $(PDDA/PAA-MoS_2/PAM-MoS_2/PEDT-PSS)_n$ in 5 mM Fe $(CN)_6^{3-}$. Scan rate: 100 mV/s. Electrolyte: 0.1 M KCl.



Figure S11. Cyclic voltammograms of bare ITO electrode and LBL films of $(PDDA/PEDT-PSS)_n$ in 5 mM Fe $(CN)_6^{3-}$. Scan rate: 100 mV/s. Electrolyte: 0.1 M KCl.



Figure S12. Nyquist plots of electrochemical impedance spectroscopy of (PDDA/ PEDT-PSS)₂, (PDDA/PEDT-PSS)₅, (PDDA/PAA-MoS₂/PEDT-PSS)₂ and (PDDA/PAA-MoS₂/PAM-MoS₂/PEDT-PSS)₅.



Figure S13. Cyclic voltammograms of bare ITO electrode and LBL films of $(PDDA/PAA-MoS_2/PAM-MoS_2/PEDT-PSS)_n$ in 4 mM H₂O₂. Scan rate: 100 mV/s. Electrolyte: 0.05 M PBS.



Figure S14. Cyclic voltammograms of bare ITO electrode and LBL films of $(PDDA/PEDT-PSS)_n$ in in 4 mM H₂O₂. Scan rate: 100 mV/s. Electrolyte: 0.05 M PBS.