Supplementary Information

Layer-by-layer assembled MoO$_2$/graphene thin film as a high-capacity and binder-free anode for lithium-ion batteries

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Fig. S1 (a) UV-visible absorption spectra of the multilayer (PDDA/GO/PDDA/PMA)$_n$ film on the quartz glass slide ($n=2$, 4, 6,...,30). (b) The peak intensity at 220 nm vs. the layer number. (c, d) SEM images for the (PDDA/GO/PDDA/PMA)$_n$ precursor.
Fig. S2 Typical EDX spectra for (a) the MoO$_3$/graphene composite obtained at 500 °C in a 5 % H$_2$/Ar atmosphere for 5 h and (b) the (PDDA/GO/PDDA/PMA)$_n$ precursor on the Ti foil obtained by the LBL process.
**Fig. S3** SEM image obtained in the EDX-mapping mode and corresponding elemental mapping of Mo, C, and O in the MoO$_2$/graphene hybrid film.
Fig. S4 TG curve for the as-formed MoO$_2$/graphene hybrid measured at a heating rate of 10 °C min$^{-1}$ in a flowing air. The weight decrease between 200–690 °C can be attributed to the comprehensive effects of the oxidation of MoO$_2$ and the combustion of graphene. The graphene content in the MoO$_2$/graphene product is evaluated to be about 19.3 wt%.
**Fig. S5** Electrochemical impedance spectrum of the hybrid MoO$_2$/graphene electrode after 1 discharge-charge cycle over the frequency range from 100 kHz to 0.1 Hz.
Fig. S6 The FE-SEM images of the film electrode after 100 discharge-charge cycling at a current density of 478 mA g$^{-1}$ in the voltage range of 0.01–3 V.
**Fig. S7** Left: FE-SEM image of MoO$_2$ product prepared by annealing PMA at 500 °C at a heating rate of 1 °C min$^{-1}$ in 5% H$_2$/Ar for 5 h; Right: the cycling performances of the as-made MoO$_2$/graphene electrode and pure MoO$_2$ electrode at a current density of 478 mA g$^{-1}$ in the voltage range of 0.01–3 V.

Pure MoO$_2$ was synthesized by annealing PMA at 500 °C at a heating rate of 1 °C min$^{-1}$ in 5% H$_2$/Ar for 5 h. The working electrode of pure MoO$_2$ was prepared by mixing 80 wt % MoO$_2$ particles, 10 wt % acetylene black (Super-P), and 10 wt % polycylylidene fluoride (PVDF).