

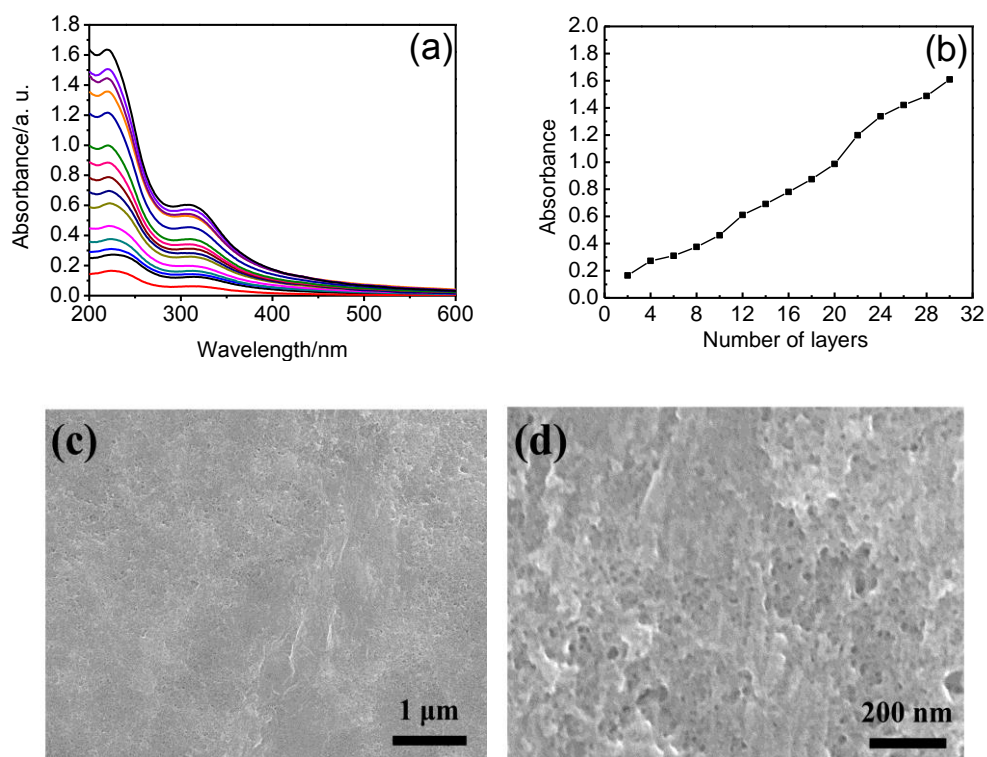
*Supplementary Information*

**Layer-by-layer assembled MoO<sub>2</sub>/graphene thin film as a high-capacity  
and binder-free anode for lithium-ion batteries**

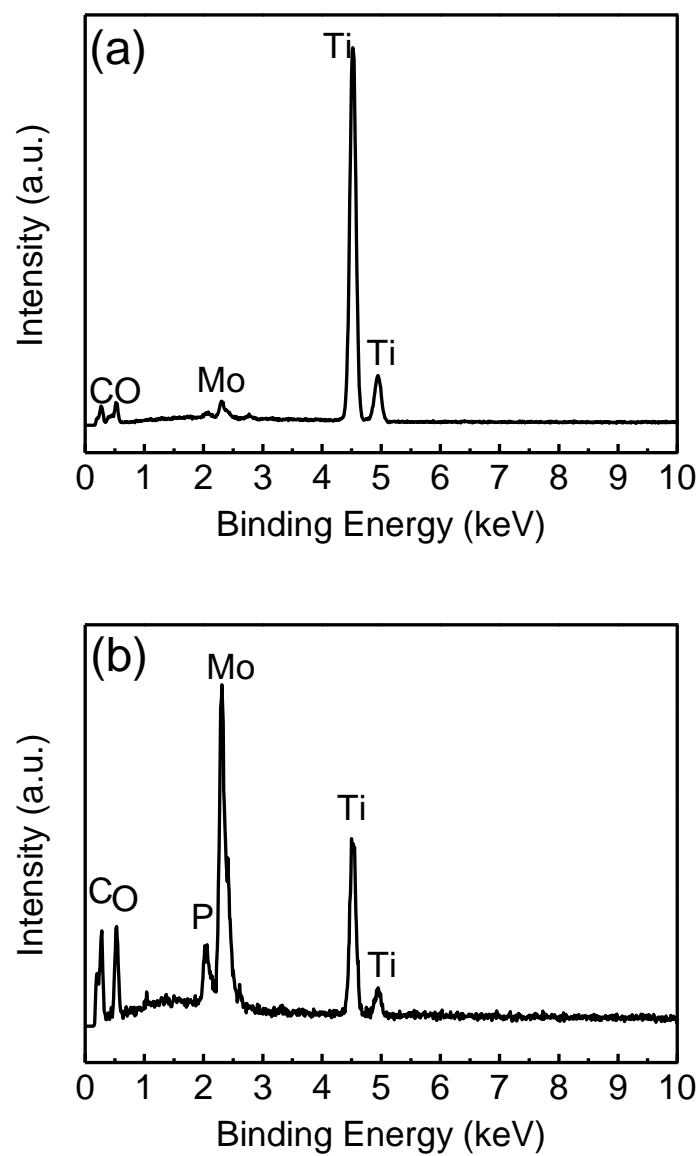
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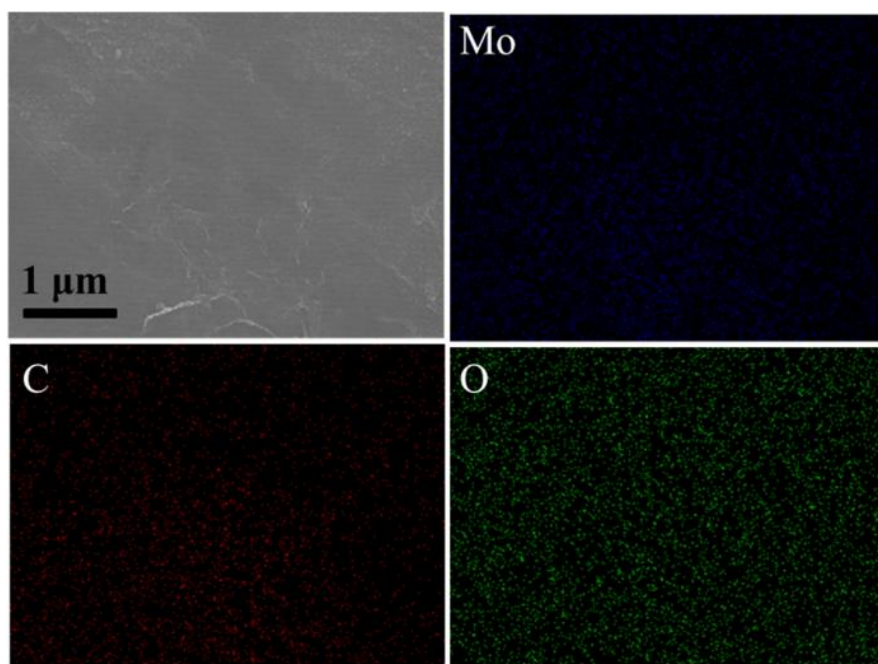
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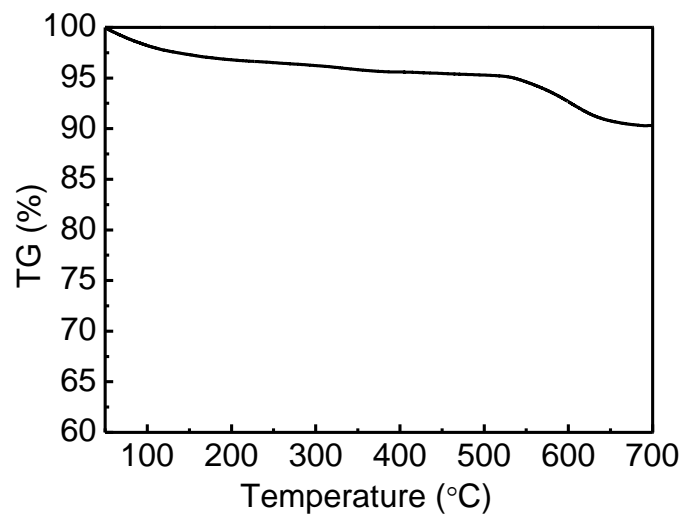
**Fig. S1** (a) UV-visible absorption spectra of the multilayer (PDDA/GO/PDDA/PMA)<sub>n</sub> film on the quartz glass slide ( $n= 2, 4, 6, \dots, 30$ ). (b) The peak intensity at 220 nm vs. the layer number. (c, d) SEM images for the (PDDA/GO/PDDA/PMA)<sub>n</sub> precursor.



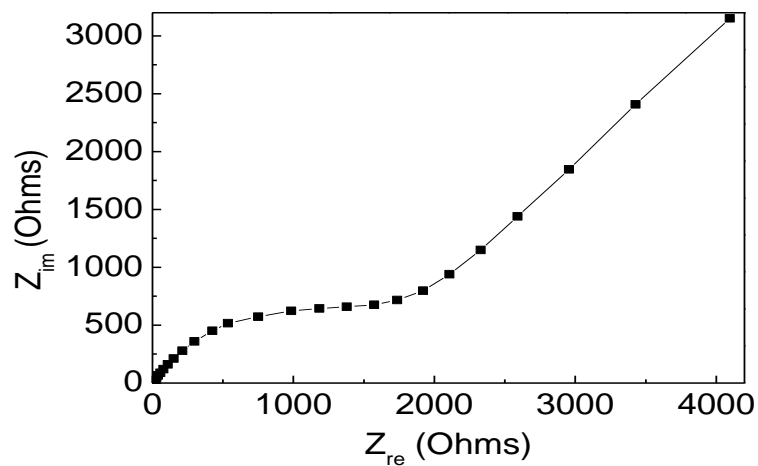
**Fig. S2** Typical EDX spectra for (a) the MoO<sub>2</sub>/graphene composite obtained at 500 °C in a 5 % H<sub>2</sub>/Ar atmosphere for 5 h and (b) the (PDDA/GO/PDDA/PMA)<sub>n</sub> precursor on the Ti foil obtained by the LBL process.



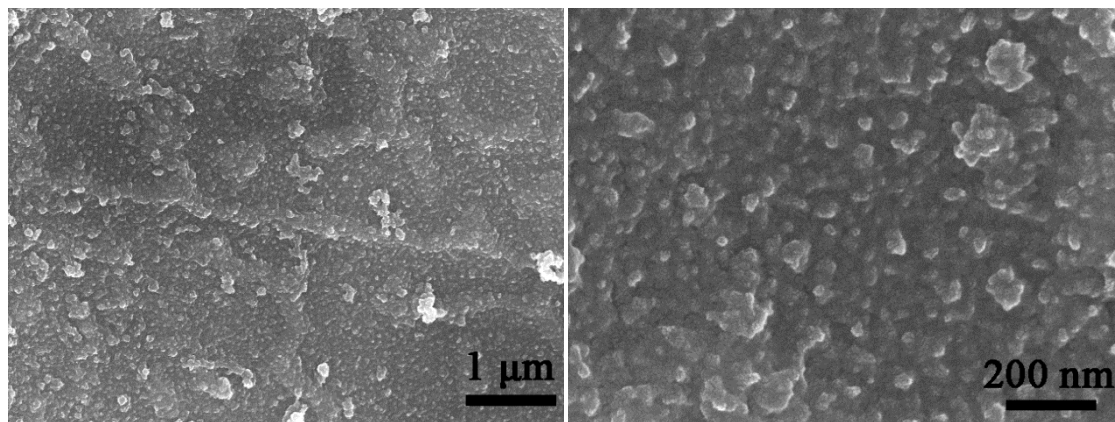
**Fig. S3** SEM image obtained in the EDX-mapping mode and corresponding elemental mapping of Mo, C, and O in the MoO<sub>2</sub>/graphene hybrid film.



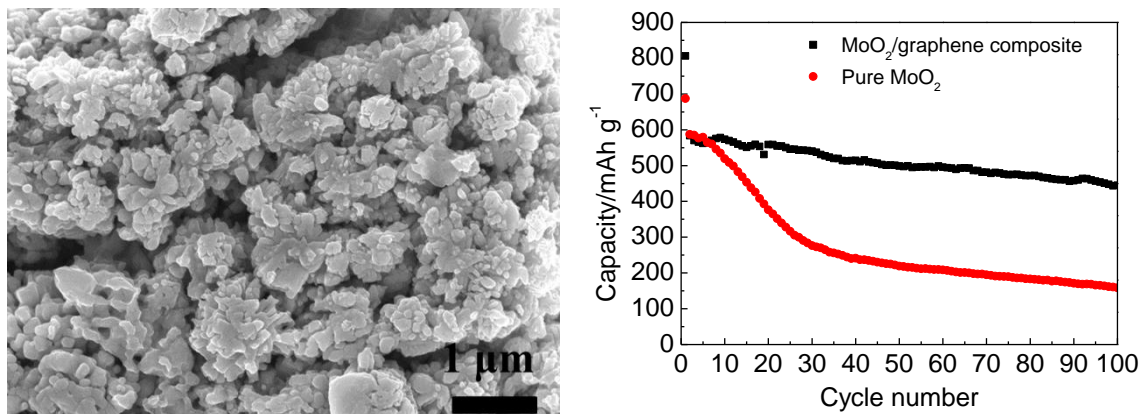
**Fig. S4** TG curve for the as-formed MoO<sub>2</sub>/graphene hybrid measured at a heating rate of 10 °C min<sup>-1</sup> in a flowing air. The weight decrease between 200–690 °C can be attributed to the comprehensive effects of the oxidation of MoO<sub>2</sub> and the combustion of graphene. The graphene content in the MoO<sub>2</sub>/graphene product is evaluated to be about 19.3 wt %.



**Fig. S5** Electrochemical impedance spectrum of the hybrid MoO<sub>2</sub>/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<sup>-1</sup> in the voltage range of 0.01–3 V.



**Fig. S7** Left: FE-SEM image of MoO<sub>2</sub> product prepared by annealing PMA at 500 °C at a heating rate of 1 °C min<sup>-1</sup> in 5% H<sub>2</sub>/Ar for 5 h; Right: the cycling performances of the as-made MoO<sub>2</sub>/graphene electrode and pure MoO<sub>2</sub> electrode at a current density of 478 mA g<sup>-1</sup> in the voltage range of 0.01–3 V.

Pure MoO<sub>2</sub> was synthesized by annealing PMA at 500 °C at a heating rate of 1 °C min<sup>-1</sup> in 5% H<sub>2</sub>/Ar for 5 h. The working electrode of pure MoO<sub>2</sub> was prepared by mixing 80 wt % MoO<sub>2</sub> particles, 10 wt % acetylene black (Super-P), and 10 wt % polycylnidene fluoride (PVDF).