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Electronic Supplementary Information <u>3D walnut-shaped TiO₂/RGO/MoO₂@Mo electrode</u> exhibiting extraordinary supercapacitor performance

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Assembly of asymmetric supercapacitor (ASC) devices

The asymmetric supercapacitor (ASC) devices were fabricated by employing as-prepared TiO₂/RGO/MoO₂@Mo and commercial active carbon (YEC-8A Fuzhou Yihuan Carbon Company) in a two-electrode configuration.

More precisely, as-obtained TiO₂/RGO/MoO₂@Mo was directly used as negative electrode. The positive electrode was fabricated by mixing active carbon (12 mg), acetylene black (1.5 mg) and PVDF (1.5 mg) at a mass ratio of 8:1:1 to obtain slurry and then coating the slurry onto carbon cloth substrate (1×1 cm²). Lastly, the positive electrode was dried at 60°C for 12 h in a vacuum oven.

The weights of active materials of electrodes were 12 mg (positive electrode) and 2.4 mg (negative electrode), respectively. The assembled ASCs devices were tested in 1 M H₂SO₄ electrolyte with a two-electrode system at room temperature about 15°C.



Fig. S1 EDS mapping of $TiO_2/RGO/MoO_2$ composites.



Fig. S2 FESEM images of TiO₂/RGO/MoO₂ composites after 5000 cycles: (a) 50 k and (b) 100 k.



Fig. S3 FESEM images of $TiO_2/RGO/MoO_2$ composites: (a, b) before and (c, d) after calcination,

(a, c) 50 k and (b, d) 100 k.



Fig. S4 XRD patterns of TiO₂/RGO/MoO₂ composites before and after calcination at 450°C.



Fig. S5 Raman spectra of TiO₂/RGO/MoO₂ composites before and after calcination at 450°C.



Fig. S6 (a) CV curves at different scan rates and (b) GCD results at various current densities for

 $TiO_2/RGO/MoO_2@Mo$ electrode after calcination at 450°C in N_2 atmosphere.



Fig. S7 (a) CV curve of AC and TiO_/RGO/MoO_2@Mo electrodes at 5 mV s^-1 in three-electrode

system. (b) CV curves of $TiO_2/RGO/MoO_2@Mo//AC$ asymmetry supercapacitors devices

collected in different cell voltage windows at 20 mV s⁻¹.



Fig. S8 Electrochemical performances of active carbon as the positive electrode.



Fig. S9 EDS in TEM of TiO₂/RGO/MoO₂ composites.