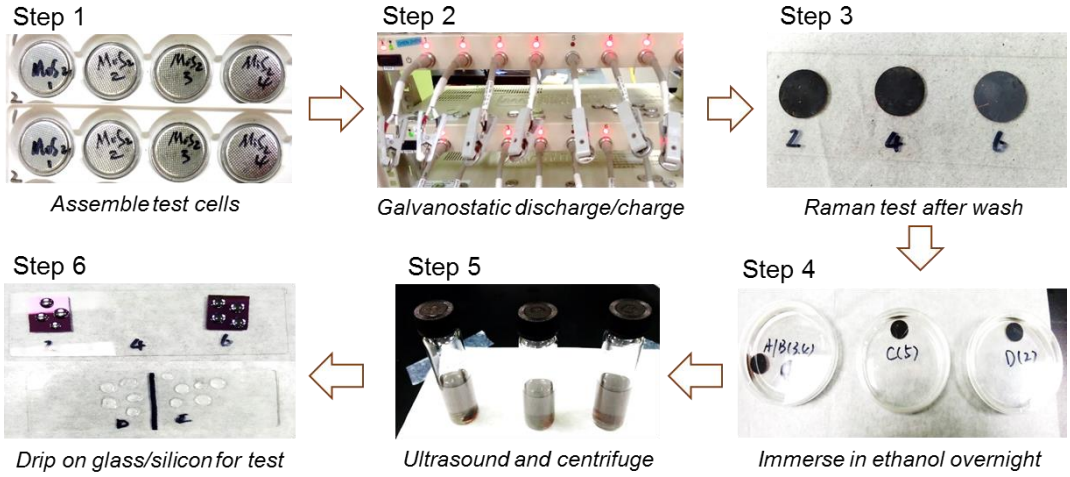

Fig. S1 Detailed experimental set-up for lithium electrochemical intercalation process of five different button cells, with MoS_2 acting as cathode and lithium plate as anode respectively.

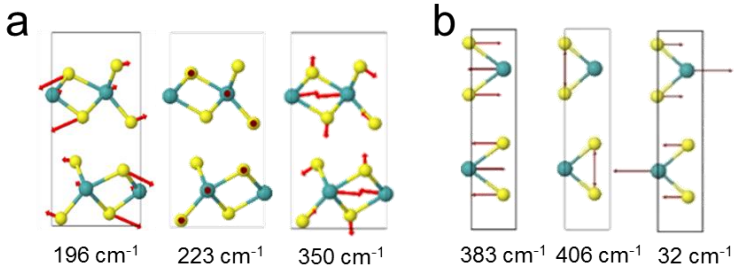

Fig. S2 The Raman vibrational diagrams of dT- (a) and 2H-MoS_2 (b) respectively. The three peaks located at 196, 223, 350 cm^{-1} in (a) are the Raman peaks of dT-MoS_2, while the peaks at 383, 406 and 32 cm^{-1} in (b) are the Raman peaks of 2H-MoS_2.

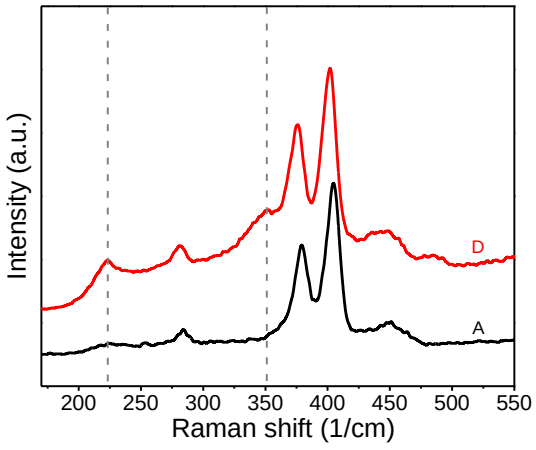

Fig. S3 Raman fingerprints of pristine MoS_2 powder and complete lithium intercalated MoS_2 with discharge potential at 0.9 V by 457 nm excitation laser. The two peaks located at 223, 350 cm^{-1} (marked with dashed lines) are the Raman feature peaks of dT MoS_2.

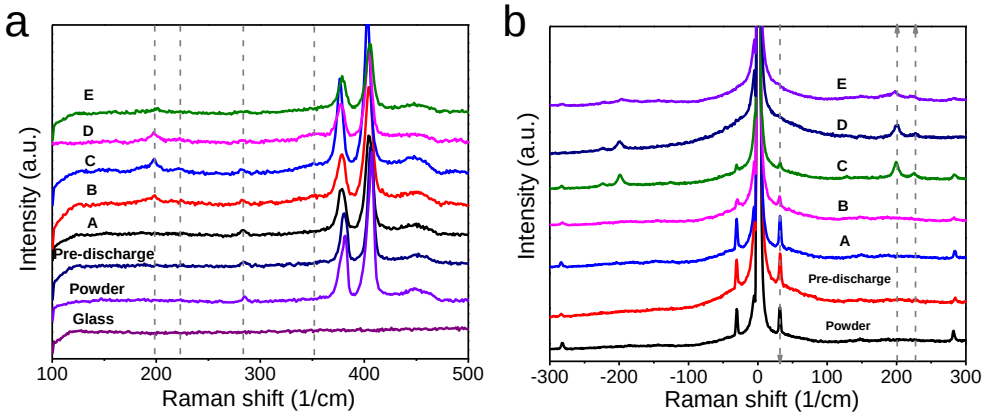

Fig. S4 High frequency (a) and ULF (b) Raman spectra of pristine MoS_2 powder and withdrawn MoS_2 cathodes in Figure 1a after ultrasonication and centrifugation four times.

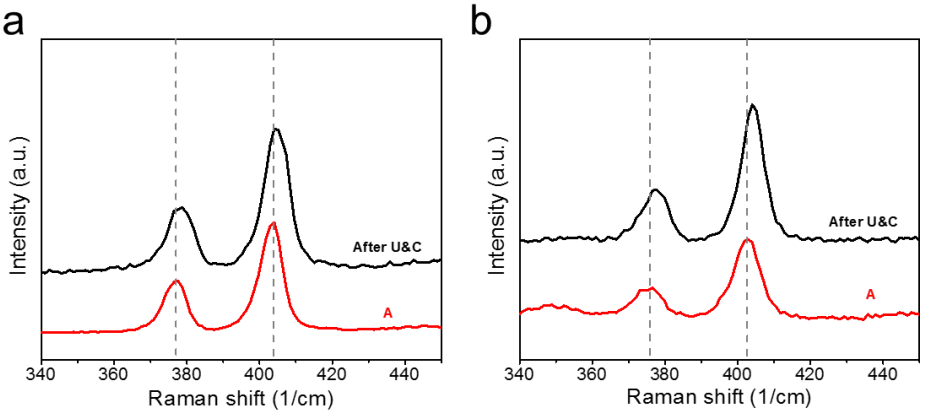

Fig. S5 Raman spectra comparison respecting $E_{2g}{}^1$ and A_{1g} modes between the just withdrawn MoS_2 cathodes after being washed and the one after ultrasonication and centrifugation four times.

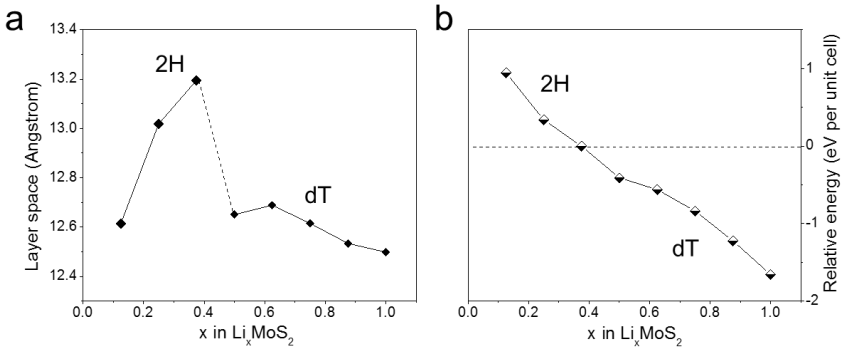

Fig. S6 Layer space evolution (a) and relative energy per unit cell (b) in Li_xMoS_2 with different Li^+ concentrations by the first-principles calculations.

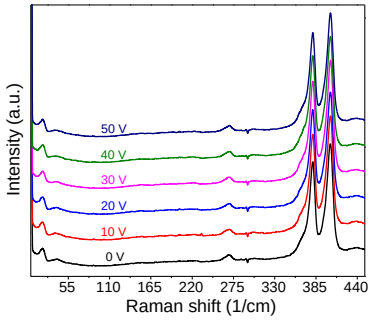

Fig. S7 Raman spectra of bilayer 2H-MoS_2 with applied gate voltages at 10 V, 20 V, 30 V, 40 V and 50 V respectively.

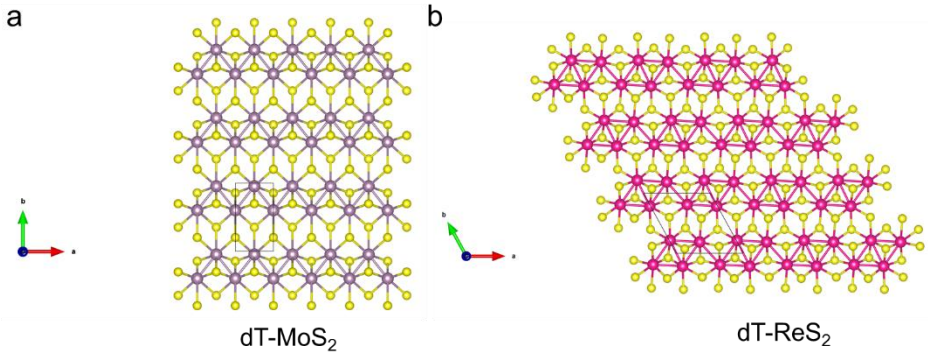

Fig. S8 Atomic structures of the top views for dT-MoS_2 and ReS_2 monolayer.