Supporting Information of

Rocksalt and Layered Metal Sulfides for Li Storage Applications: Li*Me*$_{0.5}$Ti$_{0.5}$S$_2$ (*Me* = Fe^{2+}, Mn^{2+}, and Mg^{2+})

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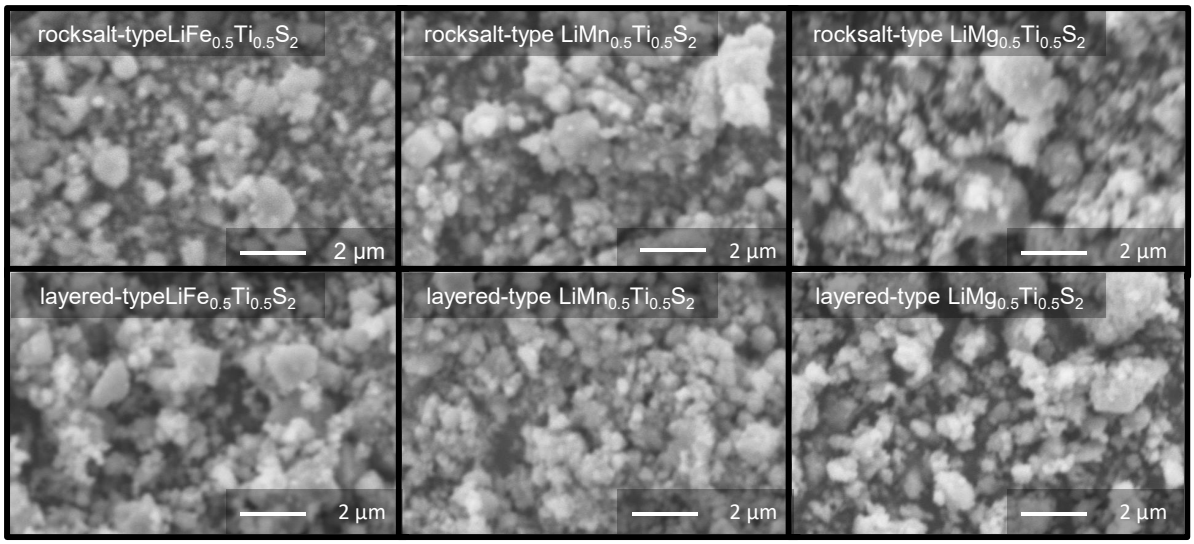
Supporting Figures



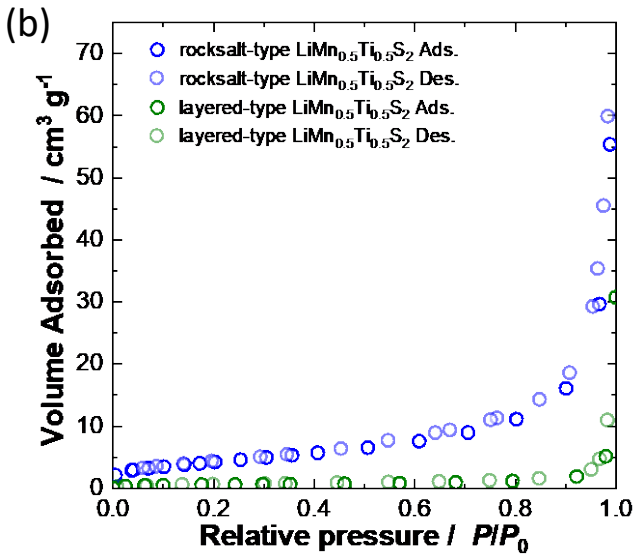


Figure S1. (a) SEM images of cation-disordered rocksalt and cation-ordered layered Li*Me*$_{0.5}$Ti$_{0.5}$S$_2$ (*Me* = Fe^{2+}, Mn^{2+}, and Mg^{2+}). (b) Results of BET specific surface area measurement for rocksalt and layered Li$_{1-x}$*Mn*$_{0.5}$Ti$_{0.5}$S$_2$ samples.

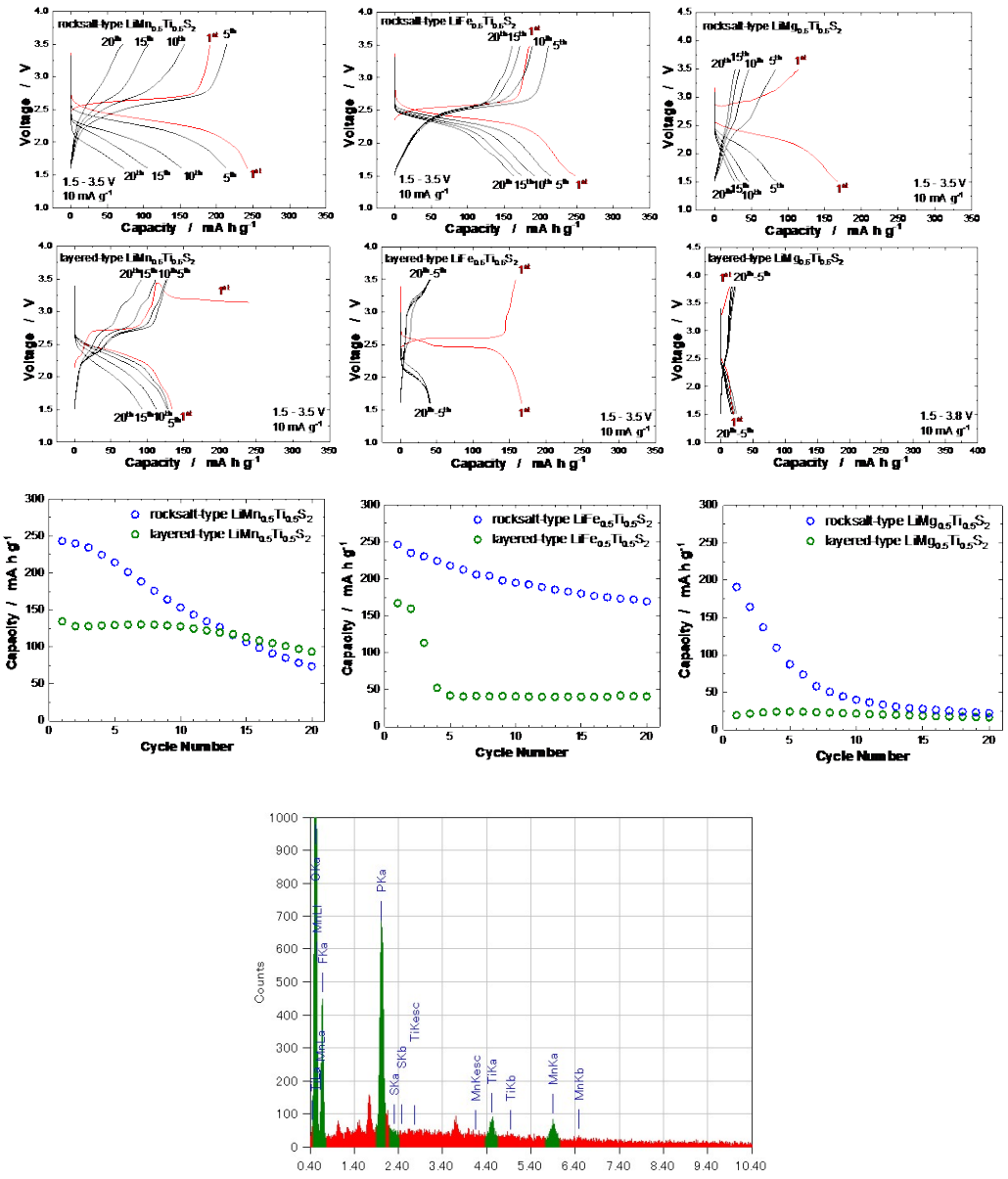


Figure S2. (a) Selected charge/discharge curves and (b) capacity retention of cation-disordered rocksalt and cation-ordered layered Li*Me*$_{0.5}$Ti$_{0.5}$S$_2$ (*Me* = Fe^{2+}, Mn^{2+}, and Mg^{2+}) at a rate of 10 mA g^{-1}. Rocksalt LiFe$_{0.5}$Ti$_{0.5}$S$_2$ shows better cyclability among the tested samples. (c) SEM/EDX spectra of the electrolyte soaked with fully charged Li$_{1-x}$Mn$_{0.5}$Ti$_{0.5}$S$_2$ at 50 ºC for 48 h. Detailed processes for the dissolution test are found in reference 23.

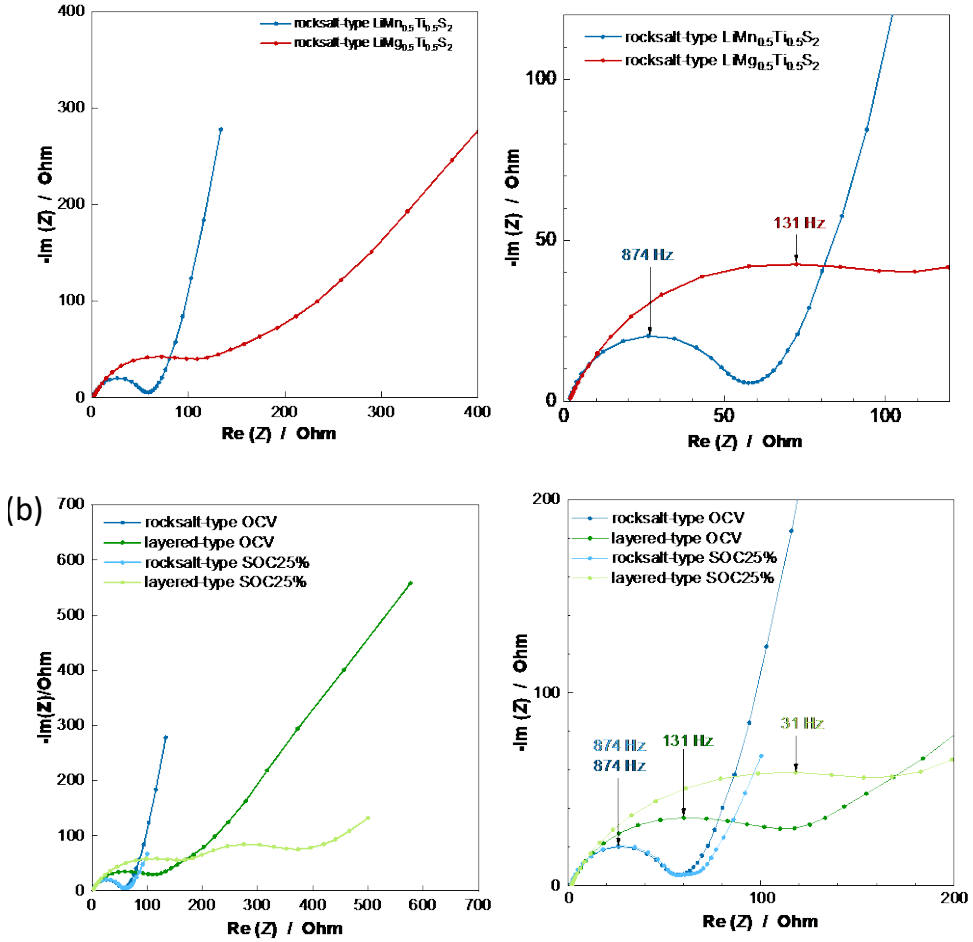


Figure S3. (a) Comparison of impedance measured at an open-circuit voltage (OCV) condition without electrochemical cycle for $LiMn_{0.5}Ti_{0.5}S_2$ and $LiMg_{0.5}Ti_{0.5}S_2$ with the rocksalt structure, and (b) impedance of rocksalt and layered $LiMn_{0.5}Ti_{0.5}S_2$ measured at OCV and 25% state of charge (SOC).

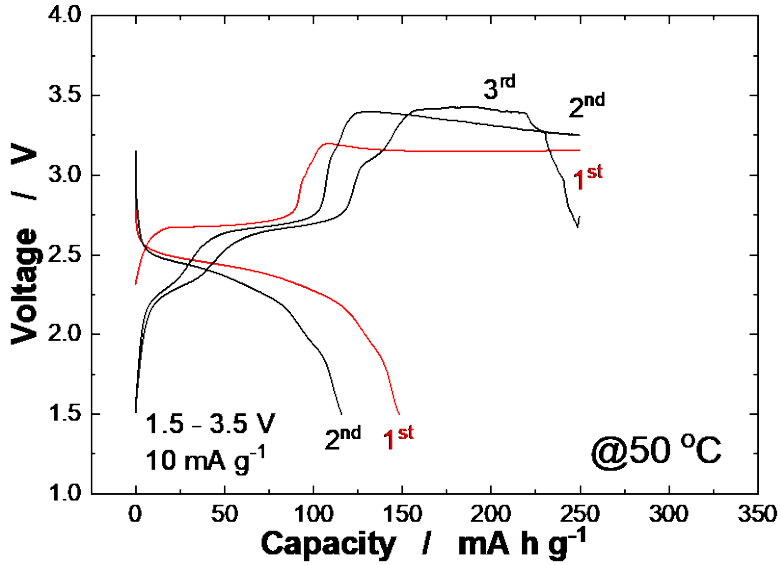


Figure S4. Charge/discharge curves of layered $LiMn_{0.5}Ti_{0.5}S_2$ at 50 °C at a rate of 10 mA g^{-1}.

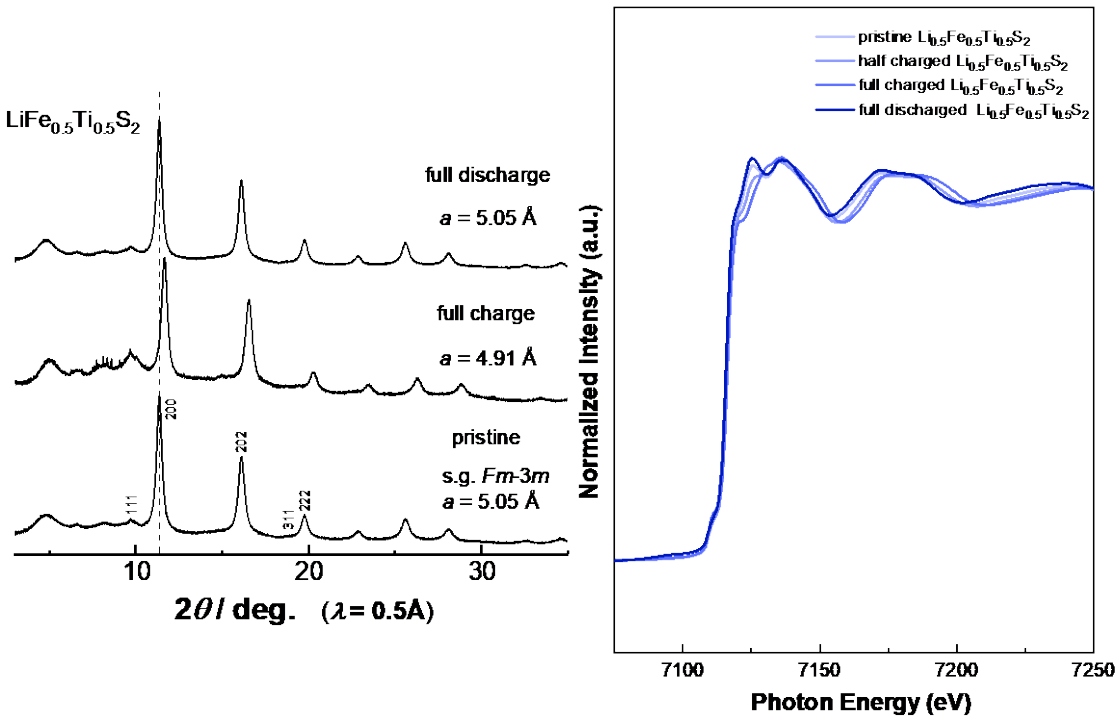


Figure S5. Structural changes of rocksalt $Li_{1-x}Fe_{0.5}Ti_{0.5}S_2$ measured by *ex-situ* XRD study. Changes in Fe K-edge XAS spectra of rocksalt $Li_{1-x}Mn_{0.5}Ti_{0.5}S_2$ on electrochemical cycles are also shown.

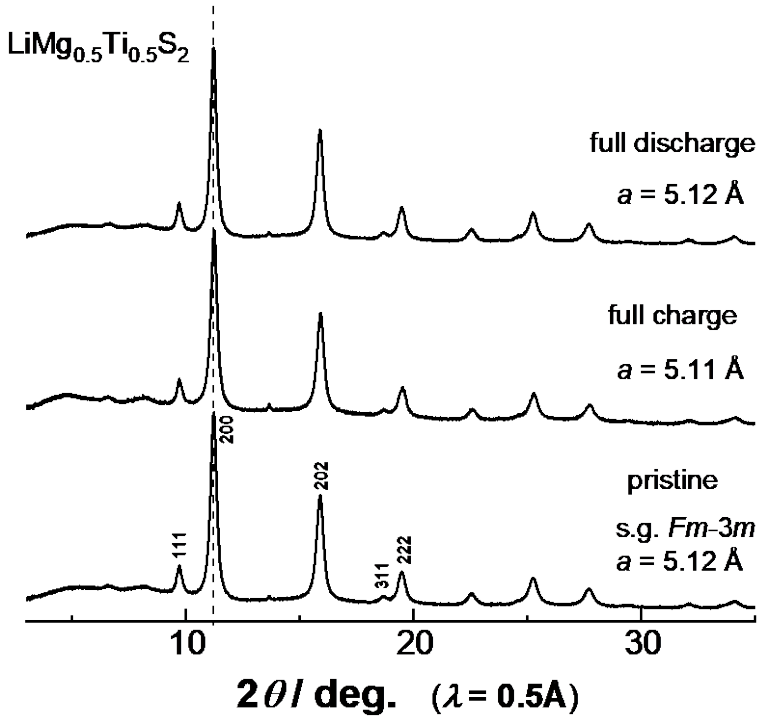


Figure S6. Structural changes of rocksalt $Li_{1-x}Mg_{0.5}Ti_{0.5}S_2$ measured by *ex-situ* XRD study. Similar to $Li_{1-x}Mn_{0.5}Ti_{0.5}S_2$, a quite small change in the unit cell volume is observed on charge/discharge cycles.