

## Long-Lifespan Lithium-Metal Batteries Obtained Using Perovskite Intercalation Layer to Stabilize Lithium Electrode

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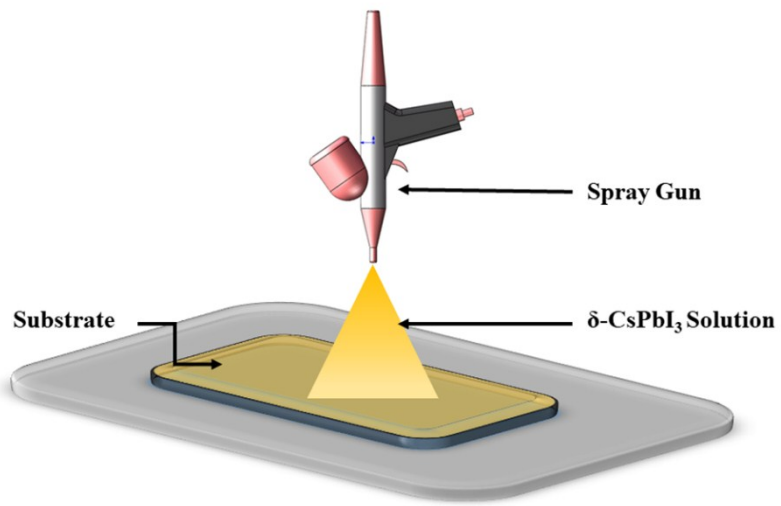
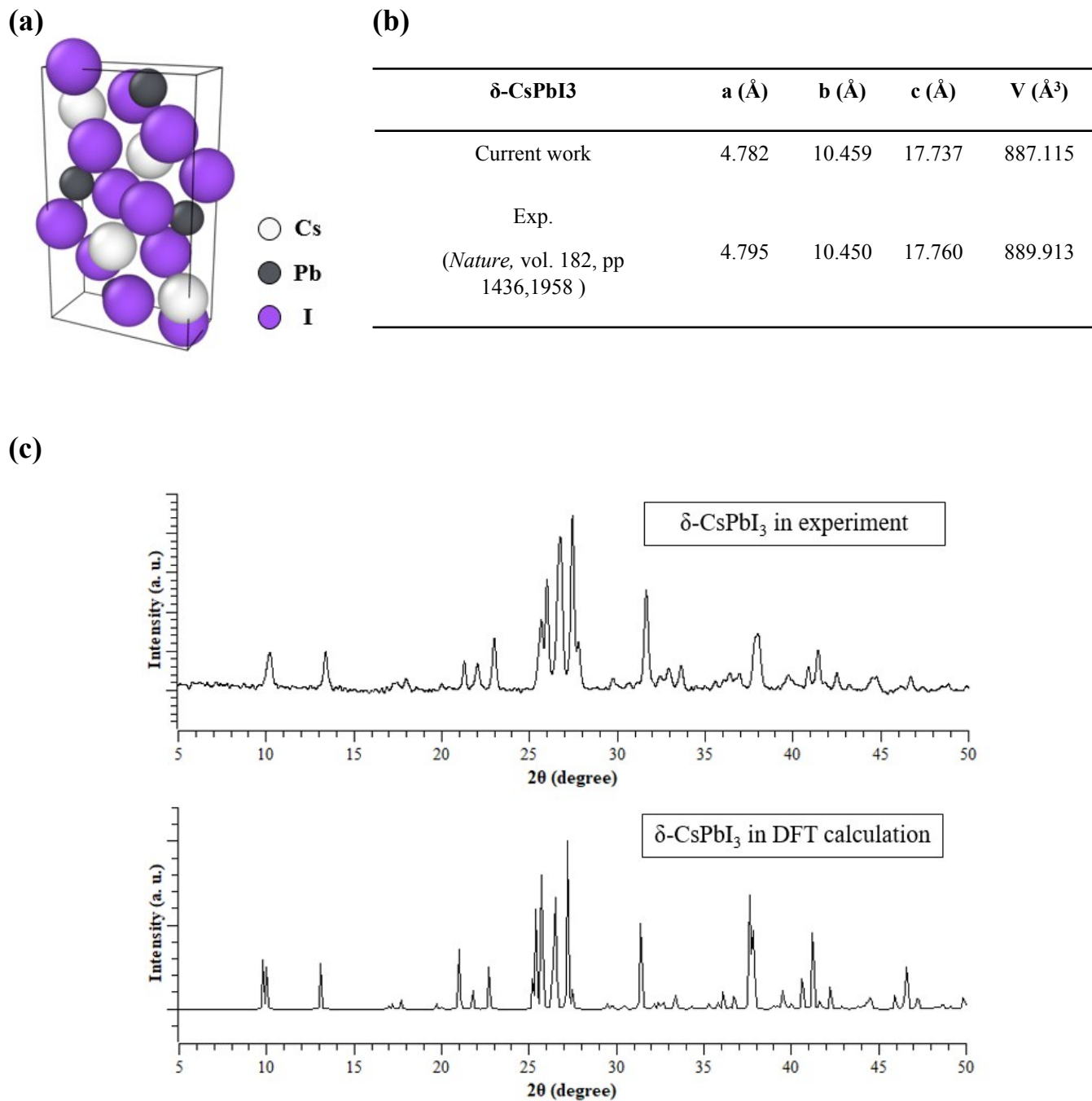
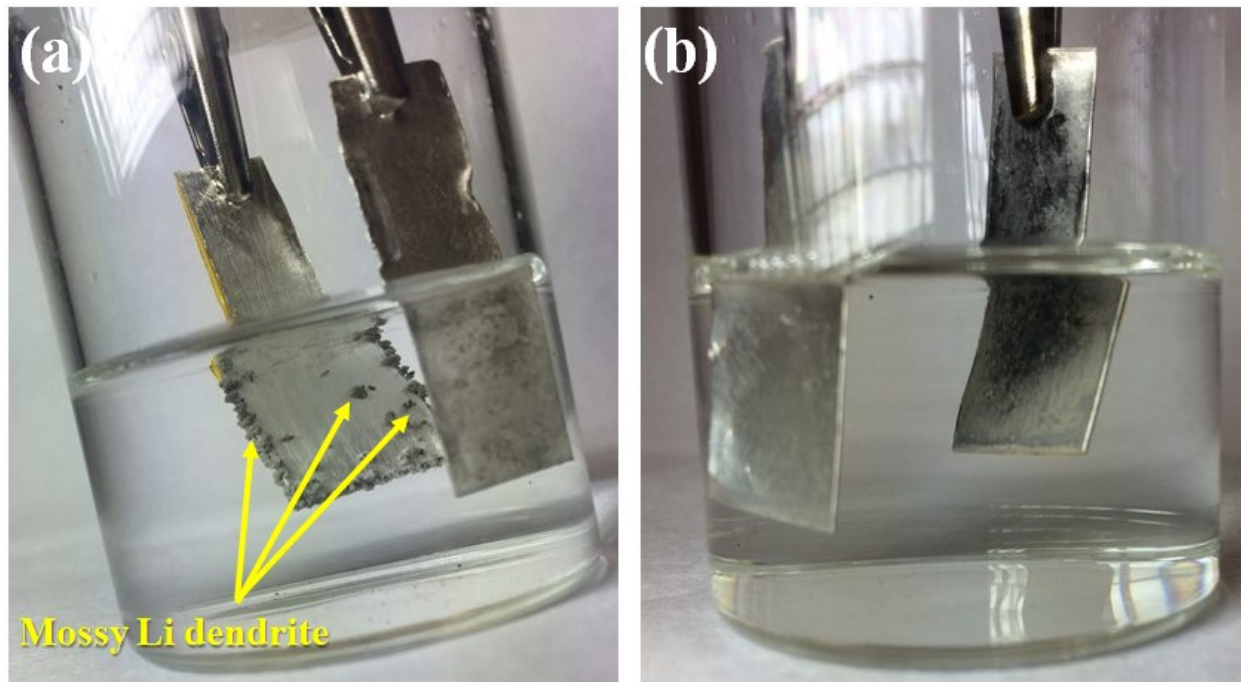


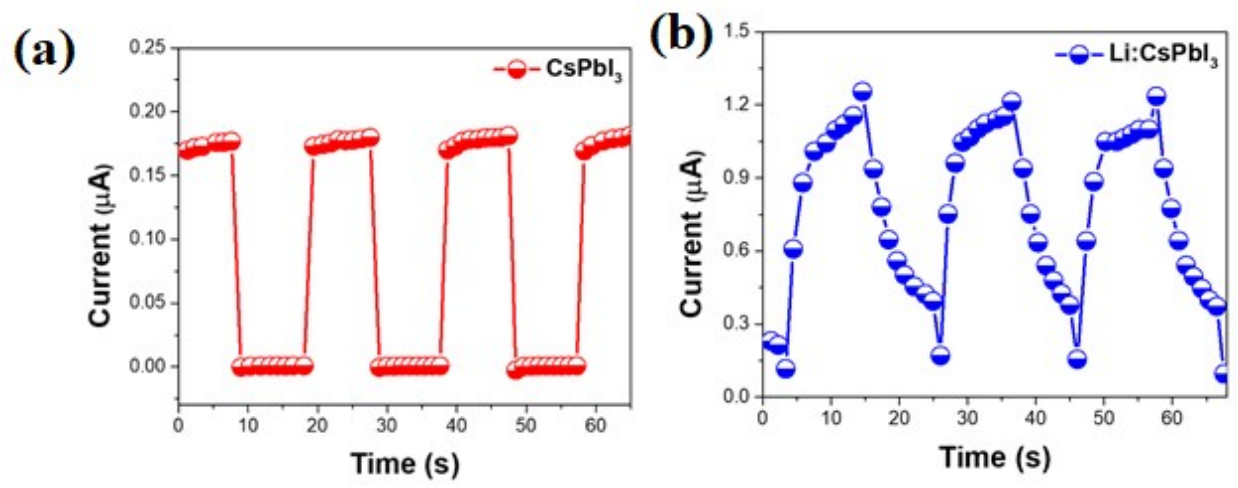
Fig. S1 Schematic representation of Spray-coating of CsPbI<sub>3</sub> using spray gun.



**Fig. S2** a) Fully optimized structure of  $\delta$ -CsPbI<sub>3</sub>. b) DFT-predicted lattice parameters and unit cell volume of  $\delta$ -CsPbI<sub>3</sub> determined in this present study, and compared with the previously reported experimental data. c) Experimentally measured and simulated of XRD patterns of  $\delta$ -CsPbI<sub>3</sub>.



**Fig. S3** Photographs of transparent cells of a) pristine Li and b)  $\delta$ -CsPbI<sub>3</sub>-coated Li after overpotential measurements.



**Fig. S4** Current vs time measurement of a) pristine  $\delta$ -CsPbI<sub>3</sub> and b) Li:CsPbI<sub>3</sub> under white light.

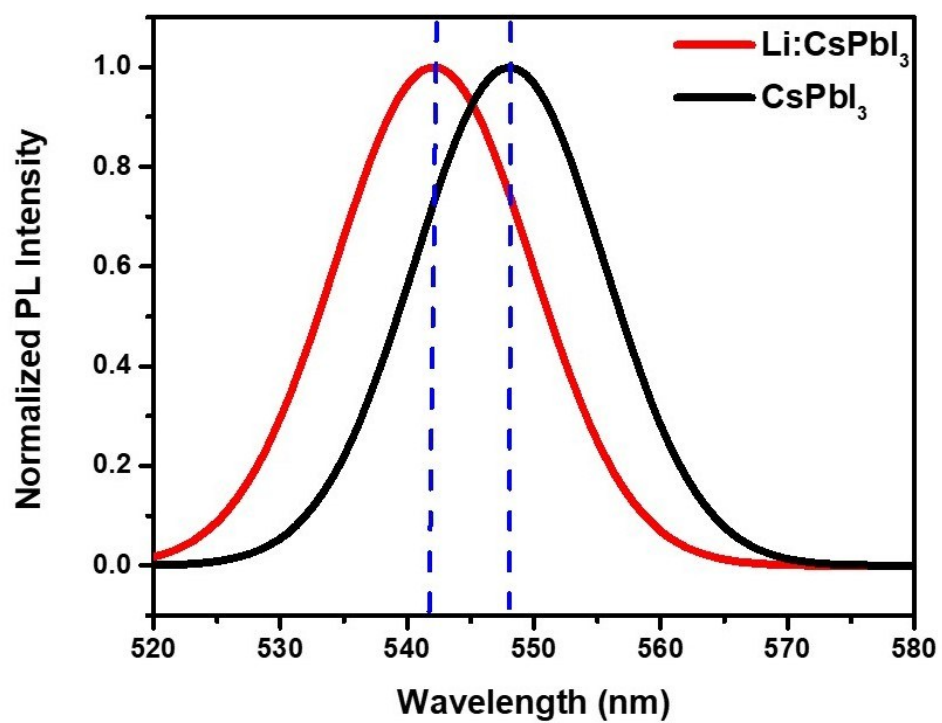
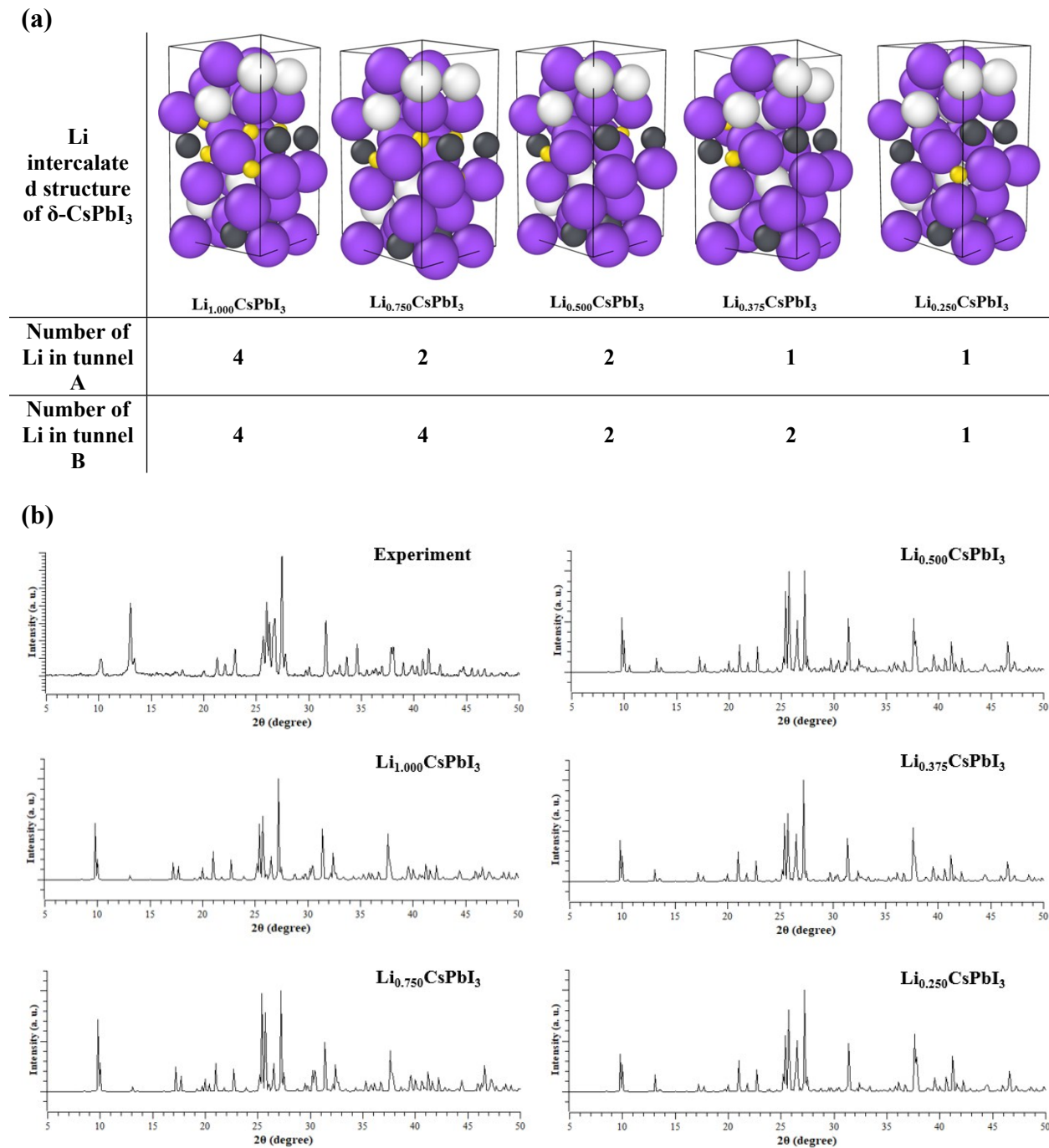
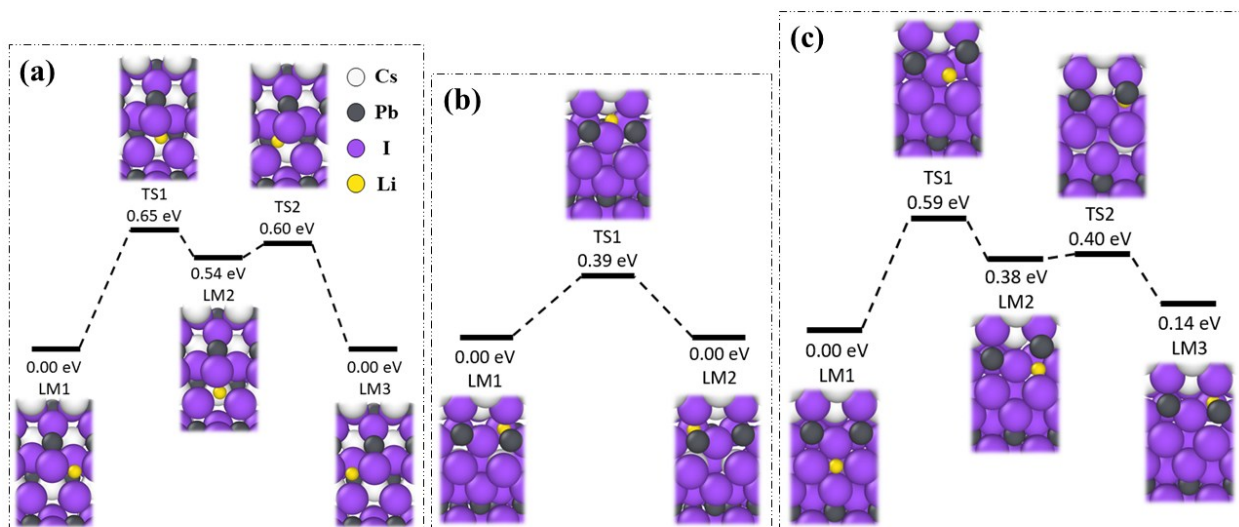


Fig. S5 PL spectra of pristine CsPbI<sub>3</sub> and Li:CsPbI<sub>3</sub>.

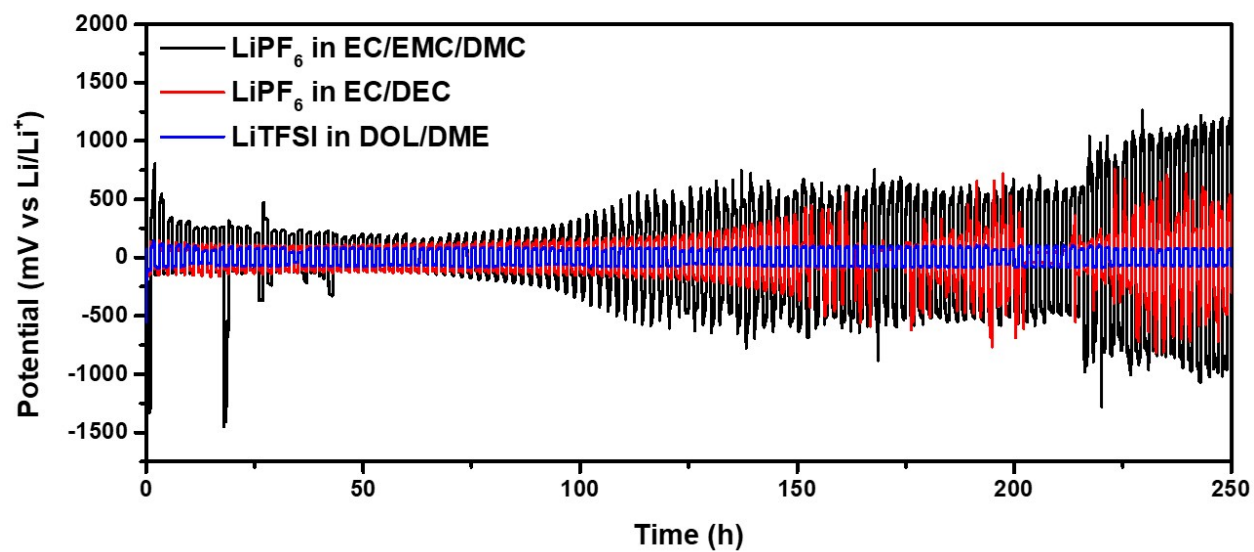


**Fig. S6** (a) Stable structures of  $\delta$ -CsPbI<sub>3</sub> featuring various ratios of intercalated Li atoms; the table lists the number of intercalated Li atoms located in tunnels A and B. (b) Experimentally measured and simulated of XRD patterns of Li-intercalated  $\delta$ -CsPbI<sub>3</sub>.

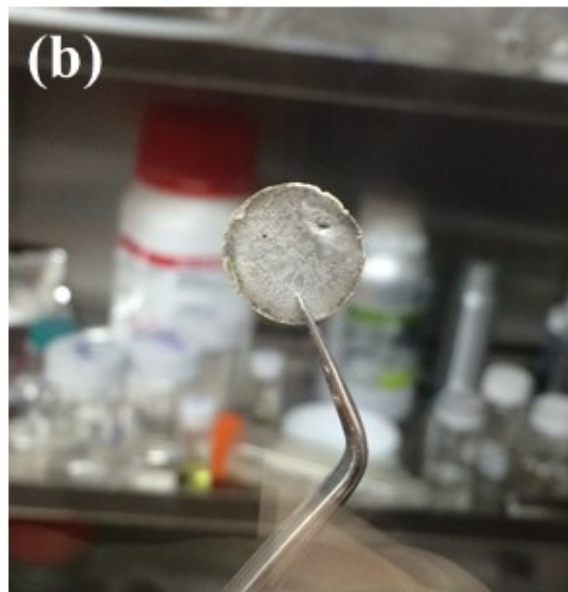
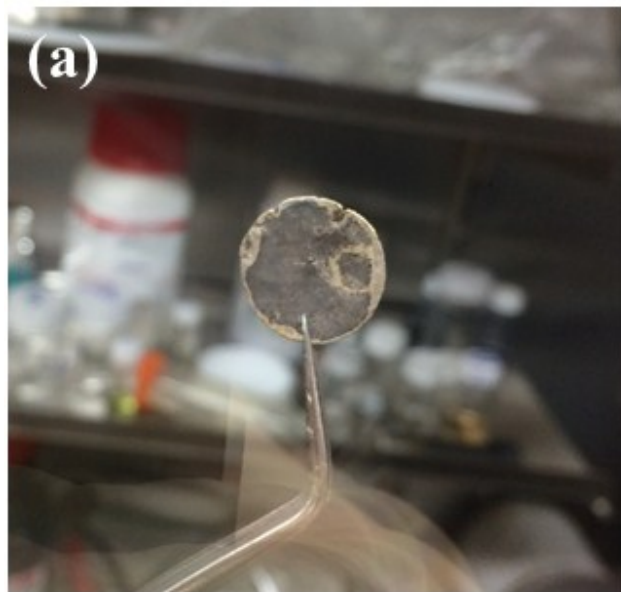


**Fig. S7** Relative energy profile of Li diffusion pathways: a) in tunnel A, b) in tunnel B, and c) from tunnel A to B.

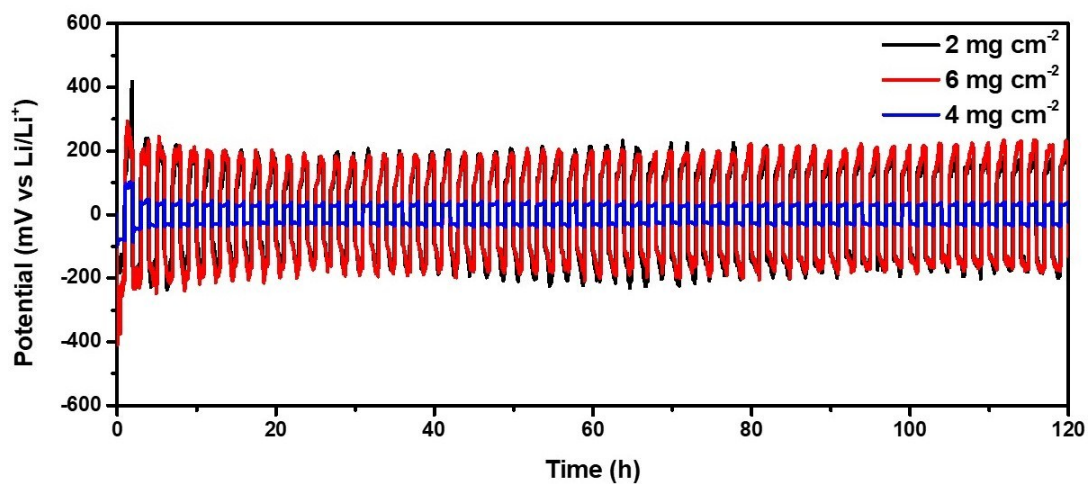




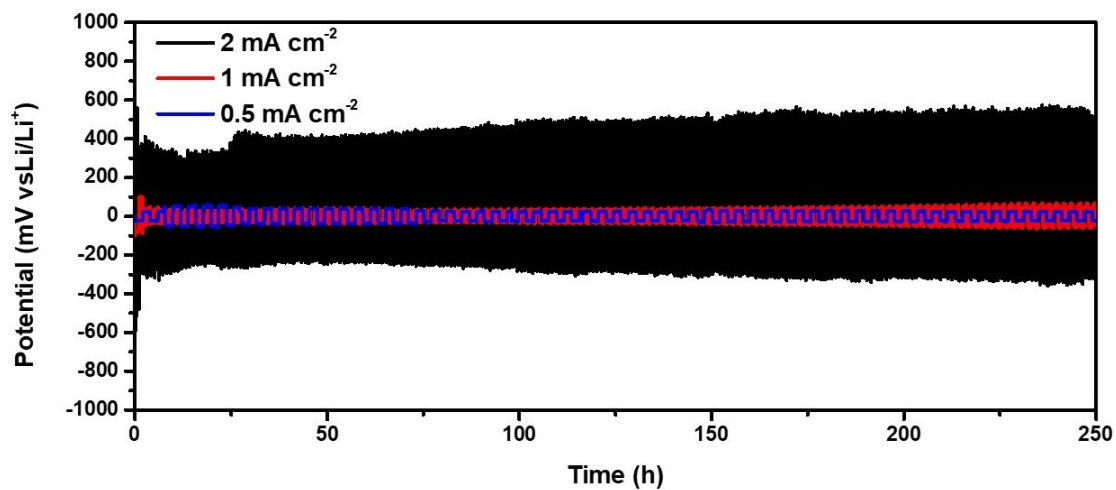
**Fig. S8** Electrochemical performance of  $\delta$ -CsPbI<sub>3</sub> in various electrolytes, recorded at a current density of 1 mA cm<sup>-2</sup> and a discharge capacity of 1 mA h cm<sup>-2</sup>.



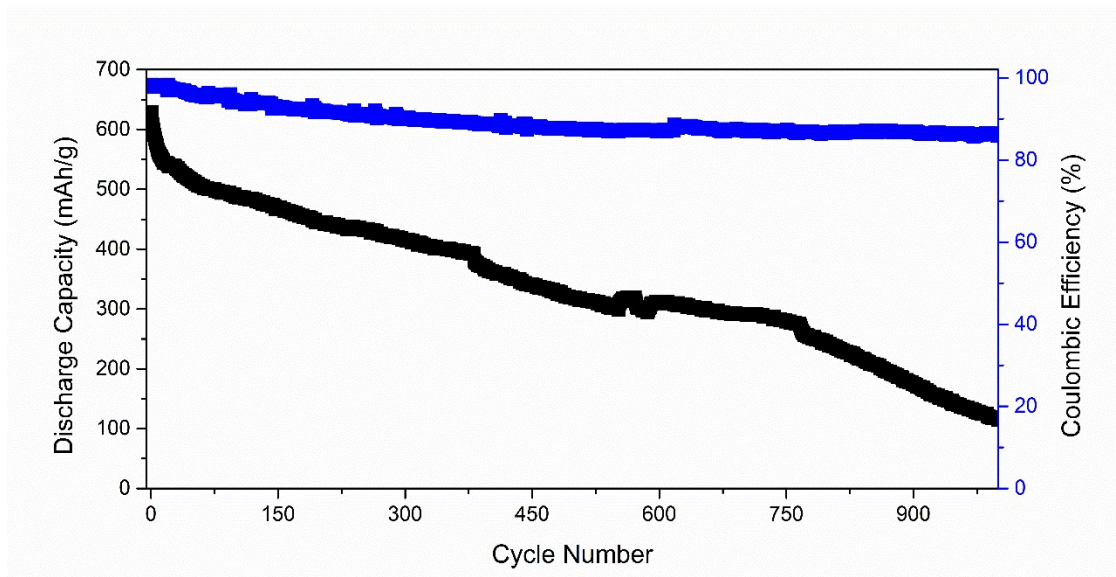
**Fig. S9** Photographs of CsPbI<sub>3</sub>-coated Li samples prepared after dissolving CsPbI<sub>3</sub> (a) in DMF/DMSO and (b) DMF.



**Fig. S10** Electrochemical performance of  $\delta$ -CsPbI<sub>3</sub> at various loadings on Li substrates, recorded at a current density of 1 mA cm<sup>-2</sup> and a discharge capacity of 1 mA h cm<sup>-2</sup>.



**Fig. S11** Electrochemical performance of Li:CsPbI<sub>3</sub>, recorded at various current densities.



**Fig. S12** Cycling performance of a Li–S battery featuring pristine Li and a separator, recorded at a 1C rate ( $1C = 175 \text{ mA h g}^{-1}$ ).