# Electrochemo-mechanical Coupling in Electrode Materials under Electrolyte-

# **Induced Interactions**

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## 1. Repeat tests

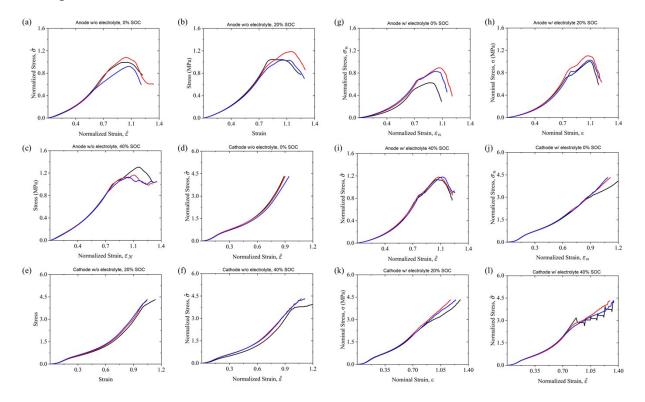
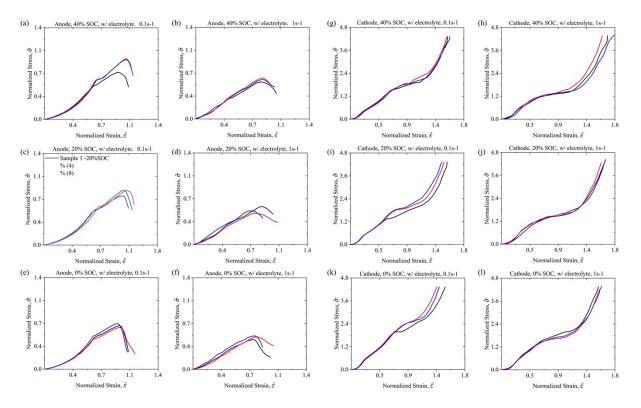


Fig. S1 (a)-(l) Compressive stress-stain curves of dry and wet electrode at various SOCs under quasi-static conditions.



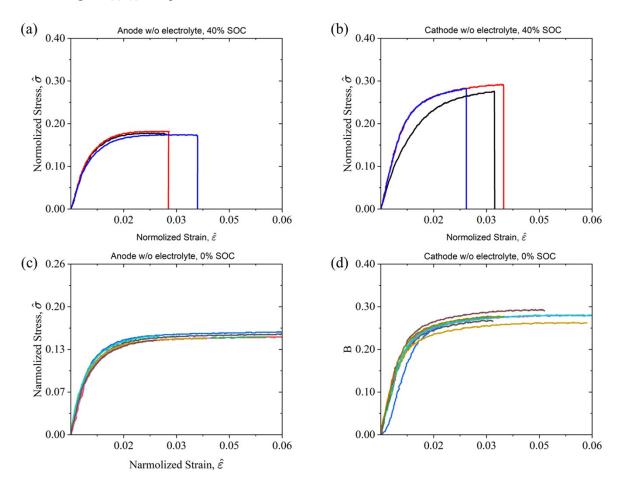


Fig. S2 (a)-(1) Compressive stress-stain curves of wet electrode at various SOCs and strain rates.

Fig. S3 (a)-(d) Tensile stress-stain curves of dry electrode at various SOCs.

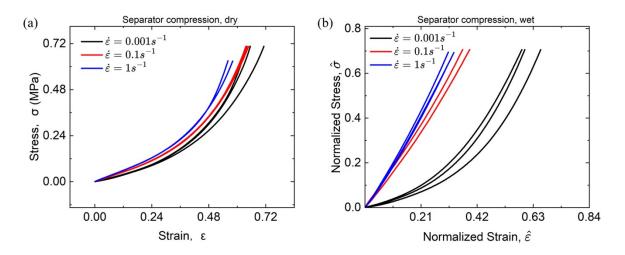


Fig. S4 (a)-(b) Compressive stress-stain curves of separator at various strain rates.

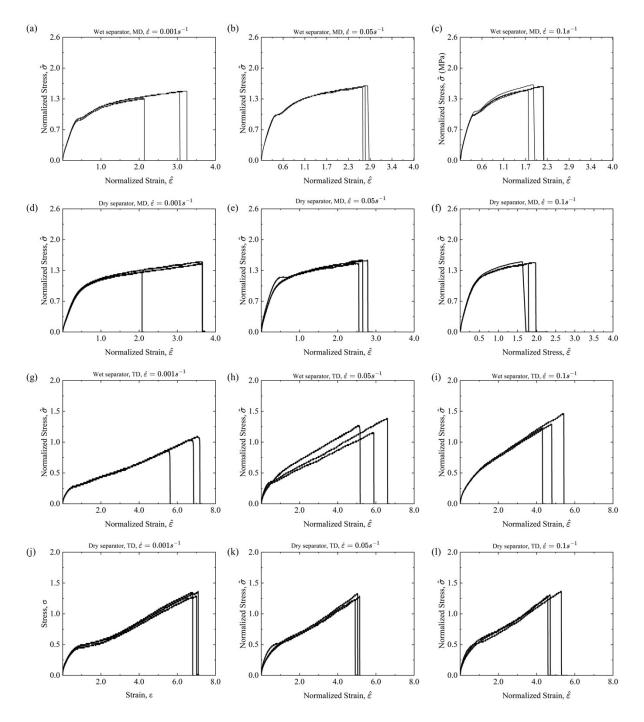
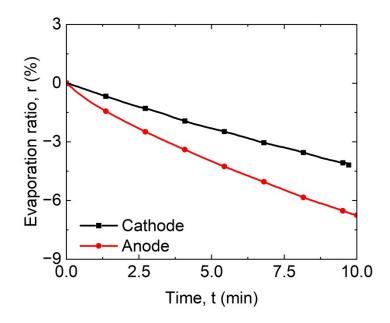
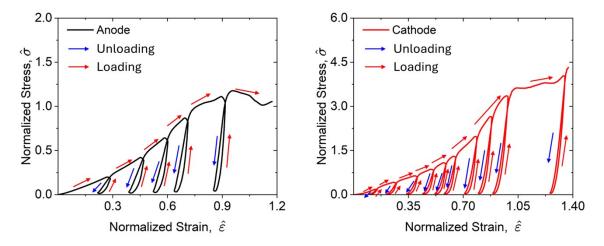


Fig. S5 (a)-(l) Tensile stress-stain curves of separator at various strain rates and two mechanical directions.

### 2. Electrolyte evaporation calibration



**Fig. S6** Cathode and anode sample electrolyte evaporation calibration. After soaking, the samples were removed from the Ziplock bag and exposed to ambient air. The mass loss observed during the resting period is attributed to electrolyte evaporation. Considering the entire testing process takes less than 2 minutes, electrolyte evaporation is limited to less than 5%.



#### 3. Loading-unloading compressive tests

Fig. S7 The cathode and anode loading-unloading tests. Unloading is performed at every force increment of  $\Delta F$ =10kN. A plastic deformation is identified at the beginning of tests, attributing to the deformation of the granular coating materials and binder relaxation.

### 4. Structural effect validation

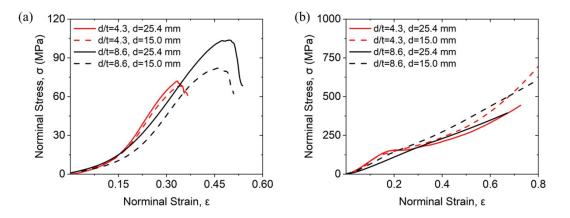


Fig. S8 Compression tests of (a) anode and (b) cathode materials were conducted with varying diameter-to-thickness ratios. A ratio of 4.3 resulted in more consistent and convergent mechanical performance across different sample diameters, indicating reduced size-related variability.