## **Solvent-free Lithium-Ion Battery Electrode with Ultrahigh Loading Using Reactive Epoxy Nano Binder**

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**Fig. S1.** Schematic diagram of a simple one-step method for the preparation of dry electrodes.

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**Fig. S2.** TEM image of EPs.



**Fig. S3.** Laser particle size scanning curve of EPs.



**Fig. S4.** (a) XPS spectra of EPs. (b) XPS C 1s spectra of EPs.



**Fig. S5.** TGA curve of EPs



**Fig. S6.** (a) Thin electrodes prepared using a 150 μm applicator. (b) Thick electrodes prepared in a single coating using a 300 μm applicator. (c) Thick electrodes prepared by double coating using a 150 μm size applicator.



**Fig. S7.** (a) Cross-sectional morphology of SEs. (b) Cross-sectional morphology of DEs.



**Fig. S8.** The SEM and elemental mapping images of the DEs.

Anode	Swelling rate $(\% )$	
	24h	48 h
<b>SEs-SBR/CMC</b>	21.29	25.16
DEs-PVDF	37.77	41.49
$DES-EPs$	11.81	14.52

**Table S1** Swelling rate of electrodes prepared by different binders in electrolyte



**Fig. S9.** Shear strength-displacement curve of EPs



Fig. S10. CV curves at a scan rate of  $0.2 \text{ mV s}^{-1}$  between 0 and  $4.3 \text{ V}$ .

## **Text S1**

The diffusivity coefficient of  $Li^+(D_{Li}^+)$  can be calculated based on the following Equation (1):

$$
D_{Li^{+}} = \frac{R^2 T^2}{2A^2 \mathbf{n}^4 F^4 C^2 \sigma^2}
$$
 (1)

Where *R* is the gas reaction constant with a calculated value of 8.314 J mol<sup>-1</sup> K<sup>-1</sup>, *T* represents the absolute thermodynamic temperature degree (298 K), *F* refers to the Faraday constant (96500 C mol<sup>-1</sup>), *A* is the area of the assembled electrode, *n* represents the number of electrons transferred in the  $Ti^{4+}/Ti^{3+}$  redox reaction (1), *C* refers to the concentration of Li<sup>+</sup> in the crystal lattice  $(4.17 \times 10^{-3} \text{ mol cm}^{-3})$ , and  $\sigma$  is the Warburg region.

Uniform dispersion of conductive agents is critical to cycling performance for the high mass loading electrodes. It can be found that MWNT in both HDEs are evenly dispersed around the active material without visible agglomeration, which confirms the universality of the dry-process in this work (Fig. S6).



**Fig. S11.** (a) Cross-sectional morphology of HDEs-EPs. (b) Cross-sectional morphology of HDEs-PVDF.



**Fig. S12.** SEM image of the self-bonding process of SBR, CMC and PVDF.



**Fig. S13.** XRD patterns of (a) fresh SBR electrode and cycled SBR electrode, (b) fresh EPs electrode and cycled EPs electrode



**Fig. S14.** XPS Li 1s spectra of (a) HDEs-EPs and (b) HDEs-PVDF after 350 cycles.



**Fig. S15.** (a) Adhesion force of the SEs, DEs-PVDF and DEs-EPs, which was characterized using (b) 180° peel-off test



**Fig. S16.** Cycling performance of HDEs-EPs-NCM811.

## **Supplementary references**

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