

## Supporting Information

### **Crosslinked, highly-conductive binder unlocks low-temperature silicon anodes for lithium-ion batteries**

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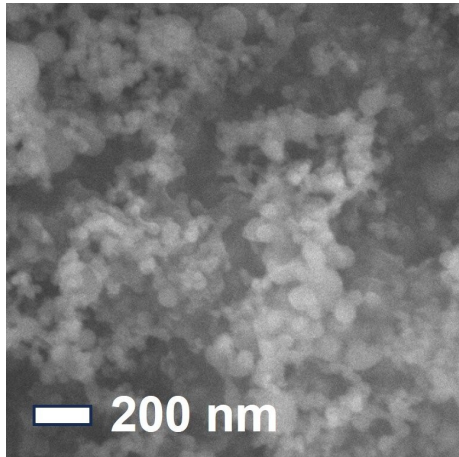
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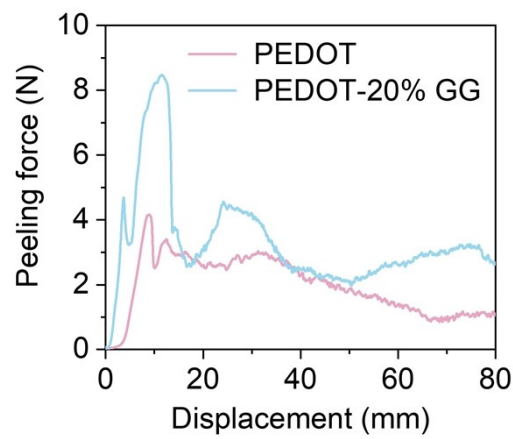
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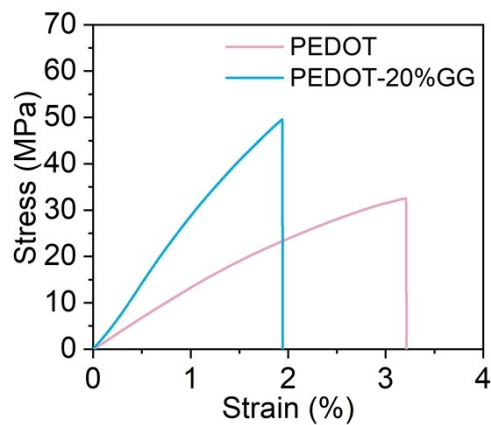
E-mail address: hlzhao@ustb.edu.cn (H. Zhao).



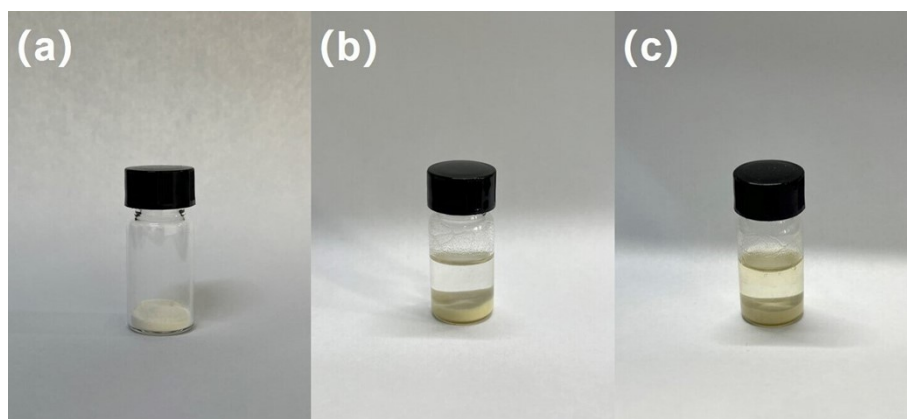
**Figure S1.** SEM image of Si power.



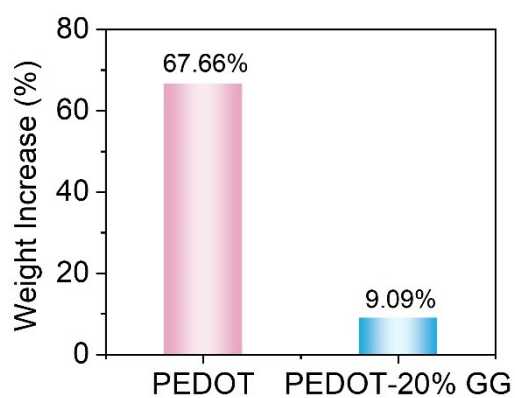
**Figure S2.** 180° peeling tests of PEDOT@Si and PEDOT-20%GG@Si electrodes on Cu foil current collectors.



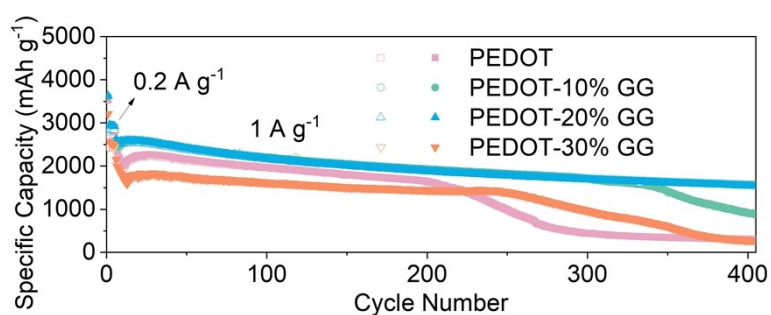
**Figure S3.** Tensile stress-strain curves of studied two binder films PEDOT and PEDOT-20%GG.



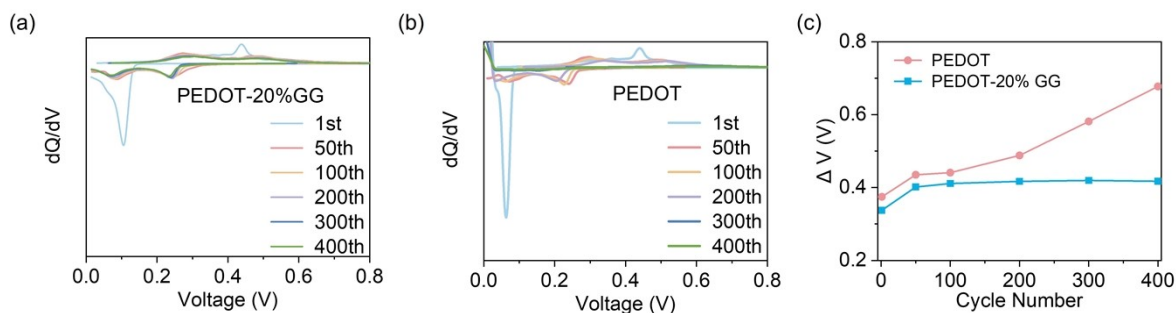
**Figure S4.** Photos of GG (a) and mixture of GG and electrolyte after 0 (b) and 24 h (c).



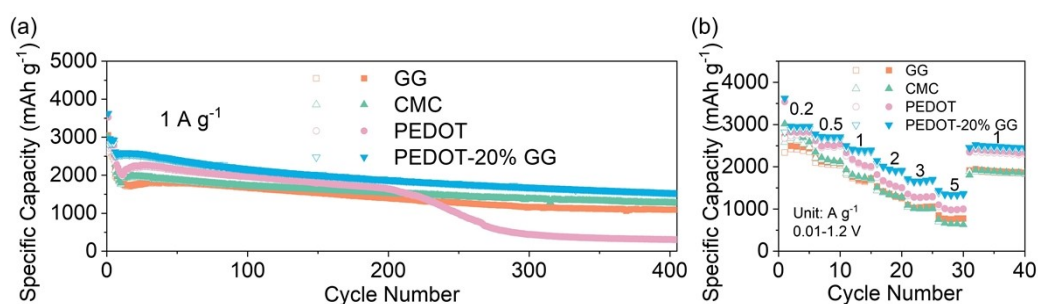
**Figure S5.** Swelling performance test of binders PEDOT and PEDOT-20%GG.



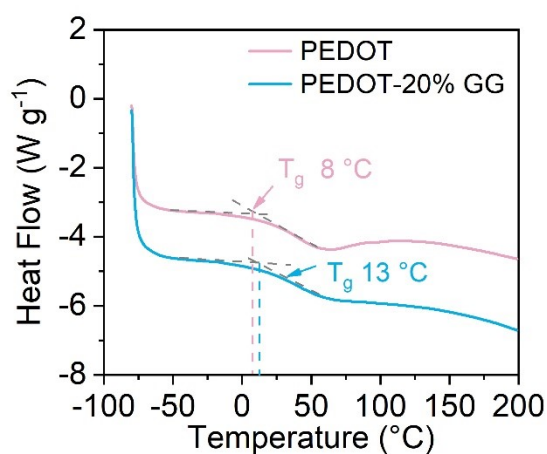
**Figure S6.** Cycling performance of PEDOT@Si, PEDOT-10%GG@Si, PEDOT-20%GG@Si, and PEDOT-30%GG@Si electrodes at a current density of 1 A g<sup>-1</sup>.



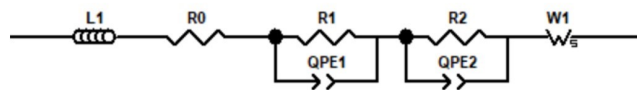
**Figure S7.** Plots of  $dQ/dV$  vs. voltage of electrodes PEDOT-20%GG@Si (a) and PEDOT@Si (b) at different cycles. (c) Voltage difference of electrodes PEDOT-20%GG@Si and PEDOT@Si.



**Figure S8.** (a) Cycling performance of electrodes GG@Si, CMC@Si, PEDOT@Si, PEDOT-20%GG@Si at a current density of  $1 \text{ A g}^{-1}$ . (b) Rate performance of electrodes GG@Si, CMC@Si, PEDOT@Si, and PEDOT-20%GG@Si.



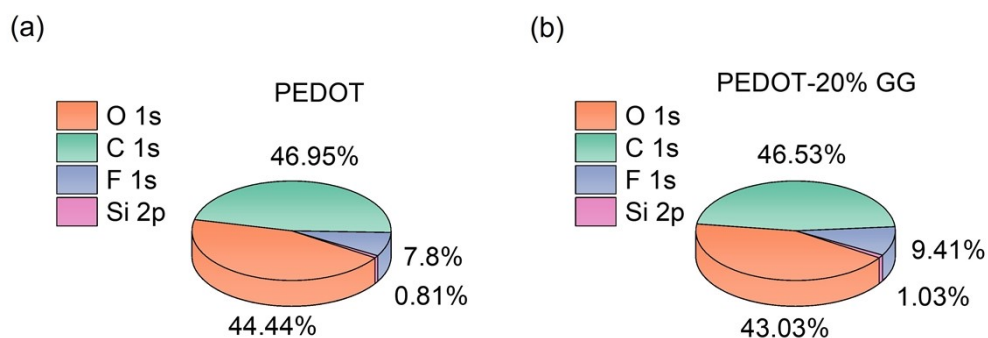
**Figure S9.** DSC curves of two binder films PEDOT and PEDOT-20%GG.



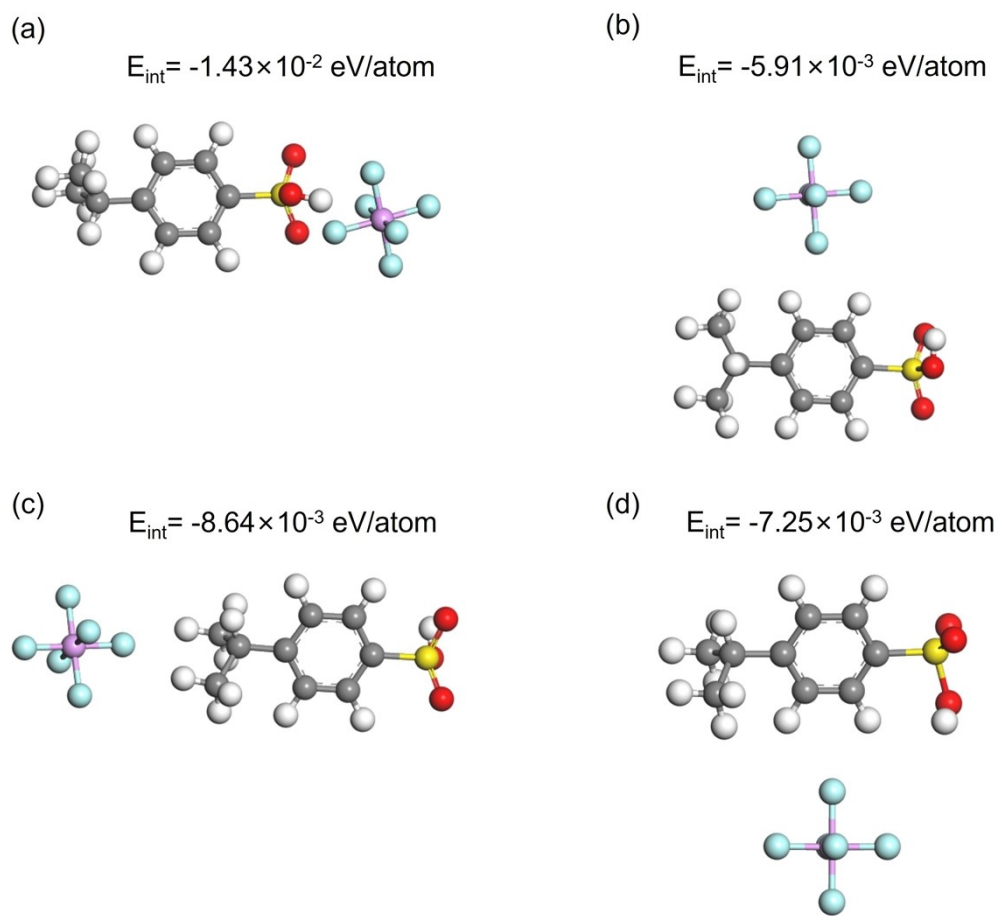
**Figure S10.** Equivalent circuit of Nyquist plot.

**Table S1.** Fitting parameters of EIS results of Si electrode with different binders at different cycles based on the data in Figure 5.

Sample	Cycle number	$R_0$ ( $\Omega$ )	$R_f$ ( $\Omega$ )	$R_{ct}$ ( $\Omega$ )
PEDOT	20	2.753	5.083	34.83
	400	7.645	11.46	45.07
PEDOT-20%GG	20	3.302	5.065	12.01
	400	5.987	6.181	28.77

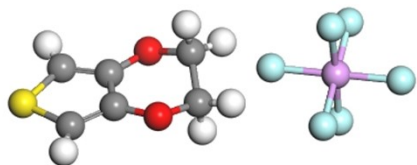


**Figure S11.** Diagram of content of each element in the SEI film of electrodes PEDOT@Si (a) and PEDOT-20%GG@Si (b) respectively, after 50 cycles, according to XPS results.

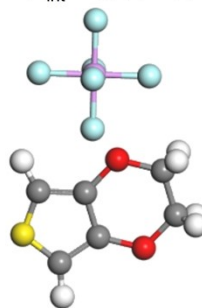


**Figure S12.** Calculation molecular structure and the obtained binding energy between PSS and  $\text{PF}_6^-$ .

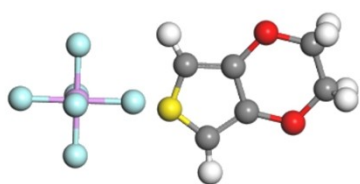
(a)  $E_{\text{int}} = -1.65 \times 10^{-2}$  eV/atom



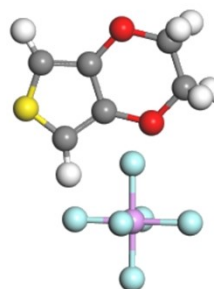
(b)  $E_{\text{int}} = -6.41 \times 10^{-3}$  eV/atom



(c)  $E_{\text{int}} = -4.05 \times 10^{-3}$  eV/atom



(d)  $E_{\text{int}} = -5.43 \times 10^{-3}$  eV/atom



**Figure S13.** Calculation molecular structure and the obtained binding energy between PEDOT and  $\text{PF}_6^-$ .