Supplementary Information

Integrating Surface Coating and Quasi-Solid Deep Eutectic Electrolytes for Enhanced Cycling of Micro-Si Anodes in Li-ion Batteries

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Fig. S1 SEM images. (a) pristine Si microparticle. (b) PD@Si microparticle.



Fig. S2 (a, b) TEM images of pristine Si particle.



Fig. S3 (a) XRD patterns of μ -Si and PD@Si powder. (b) Raman spectra of μ -Si and PD@Si powder.



Fig. S4 Electrochemical property of PD@Si anode. (a) CV curves of PD@Si anode in liquid electrolyte. (b) CV curves of PD@Si anode in gel electrolyte.



Fig. S5 Voltage profiles. (a) Pristine Si anode in liquid electrolyte. (b) Pristine Si anode in gel electrolyte.



Fig. S6 Cycling performance at 0.5 A/g. (a) Pristine Si anode in liquid electrolyte (black) and gel electrolyte (red). (b) Comparison cycling performance of pristine Si (blue) and PD@Si anodes (red) in gel electrolyte.



Fig. S7 The rate performance of pristine Si and PD@Si anodes in gel electrolyte.



Fig. S8 SEM images. (a) Pristine Si electrode surface before cycling. (b) PD@Si electrode surface before cycling.



Fig. S9 Morphology evolution. (a) Pristine Si anode surface after cycling in liquid electrolyte. (b) Pristine Si anode surface after cycling in gel electrolyte.

| Si anode | Current density | Capacity (mAh/g) after cycling | Cycles | References |
|--|--------------------|-----------------------------------|--------|------------|
| SiMP | 0.1C | 1700 | 100 | [S1] |
| Si@MOF | 0.2 A/g | 1442 | 50 | [S2] |
| PANa _{0.8} Fe _{0.01} /Si-MP/CB | 0.5C | 1386.3 | 400 | [S3] |
| Micro-sized bulk porous Si | 0.5 A/g | 1250 | 100 | [S4] |
| Submicron Si particle | - | 1192 | 100 | [S5] |
| PD@Si | 1 A/g | 1000 | 100 | This work |
| Raspberry-like YS Si/C | 0.2 A/g | 1064 | 250 | [S6] |
| SiO | - | 1068 | 150 | [S7] |
| Si-coated VACNF | 2.6 A/g | 1050 | 120 | [S8] |
| Si@SiO2@LPO@C | 0.5 A/g | 1012.4 | 200 | [26] |
| DP-Si-1.0 | 0.5 A/g | 935.9 | 100 | [S9] |
| SiO _x /C-6 | 0.1 A/g | 926.5 | 100 | [S10] |
| Mxene-Si-CNT | 2 A/g | 841 | 200 | [S11] |
| Porous SiMP | 0.5 A/g | 796 | 100 | [S12] |
| SiFS/G@C | 1 A/g | 729.1 | 100 | [S13] |
| SiNP/DPA | 1C | 750 | 100 | [S14] |
| PD-coating Si/C | 0.1C | 560 | 100 | [S15] |

Table S1. Table of comparison LIBs performcane of our work and recently reported composite Si nanoparticles and μ -Si-based anodes.

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