**Supplementary Materials**

**Bubble wrap-like carbon-coated rattle-type silica@silicon nanoparticles by surface-protected etching as hybrid anodes for lithium-ion batteries**

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**Figure S1.** FE-SEM image of (a) pure Si nanoparticles. (b) FT-IR result comparing pure Si nanoparticles and Si-OH after piranha solution pre-treatment.



**Figure S2.** (a) Formation of siloxane networks from piranha-treated Si-OH using APTES. (b) Hydrolysis and (c) condensation reaction mechanism of TEOS showing formation of siloxane bridges.



**Figure S3.** XRD patterns of (a) Si and (b) TEOS-derived SiO2@Si and APTES/TEOS-derived SiO2@Si.

Chart, histogram

Description automatically generated

**Figure S4.** XRD reference peaks for face-centered cubic Si crystal (Reference code 98-065-2265)

**Peak list**

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| No. | h | k | l | d [Å] | 2Theta[deg] | I [%] |
| 1 | 1 | 1 | 1 | 3.13559 | 28.442 | 100.0 |
| 2 | 0 | 2 | 2 | 1.92015 | 47.302 | 65.0 |
| 3 | 1 | 1 | 3 | 1.63751 | 56.122 | 37.8 |
| 4 | 2 | 2 | 2 | 1.56779 | 58.856 | 0.0 |
| 5 | 0 | 0 | 4 | 1.35775 | 69.129 | 9.8 |
| 6 | 1 | 3 | 3 | 1.24596 | 76.375 | 14.4 |
| 7 | 2 | 2 | 4 | 1.10860 | 88.029 | 19.9 |



**Figure S5.** TEM images showing carbon crosslinking from PEI and PDA carbon coating efficiency in (a, b) composite samples prepared via traditional SiO2 sol-gel synthesis reaction and carbon coating at room temperature and (c, d) composite samples prepared via hydrothermal treatment at 140 ºC for 24 h.



**Figure S6.** Illustration of the two possible reaction mechanisms showing the formation of PDA-PEI carbon crosslinks.



**Figure S7.** Raman peak fitting of core shell PDA@SiO2@Si composite sample.



**Figure S8.** Raman peak fitting of yolk shell PDA-PEI@SiO2@Si composite sample.



**Figure S9.** Raman peak fitting of yolk shell PDA-PEI@PVP-SiO2@Si composite sample.



**Figure S10.** Raman peak fitting of core shell PDA-PEI@SiO2@Si composite sample.



**Figure S11.** CV scans of core shell PDA-PEI@TEOS-SiO2@Si composite fabricated via RT using TEOS as precursor solution.



**Figure S12.** Galvanostic charge and discharge profiles of core shell PDA@SiO2@Si (a), core shell PDA-PEI@SiO2@Si (b), core shell PDA-PEI@PVP-SiO2@Si (c), and yolk shell PDA-PEI@SiO2@Si (d) composite samples.

**Table S1**. The ID/IG values of the samples calculated by the ratio of the D band peak area to the G band peak area using Gaussian-Lorentzian curve fitting model.

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| **Sample** | **D band** | | | **G band** | | | **ID/IG** |
| **A** | **I** | **FWHM** | **A** | **I** | **FWHM** |
| Core shell PDA@SiO2@Si | 15244.277 | 1347.0 | 51.480 | 10940.822 | 1597.0 | 95.0 | 0.84 |
| Yolk shell PDA-PEI@SiO2@Si | 29685.0 | 1350.0 | 420.0 | 11177.740 | 1585.0 | 120.0 | 0.85 |
| Yolk shell PDA-PEI@PVP-SiO2@Si | 25087.054 | 1361.0 | 190.0 | 11283.708 | 1586.0 | 100.0 | 0.86 |
| Core shell PDA-PEI@SiO2@Si | 17354.846 | 1348.0 | 180.0 | 9529.654 | 1581.0 | 100.0 | 0.85 |