Supporting Information

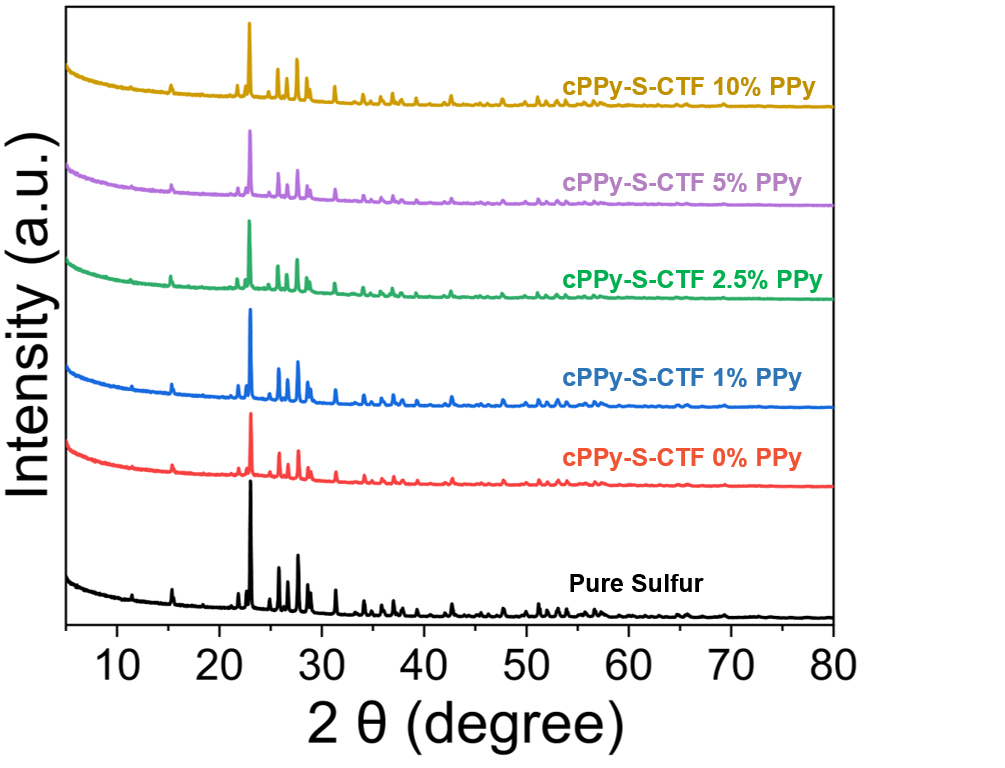
**Covalent Triazine Framework Incorporating Charged Polypyrrole Channels for High Performance Lithium-Sulfur Batteries**

Jiheon Kim,†,‡ Ahmed Elabd,§,‡ Sung-Yoon Chung,\*, † Ali Coskun,\*, § Jang Wook Choi\*, ҩ

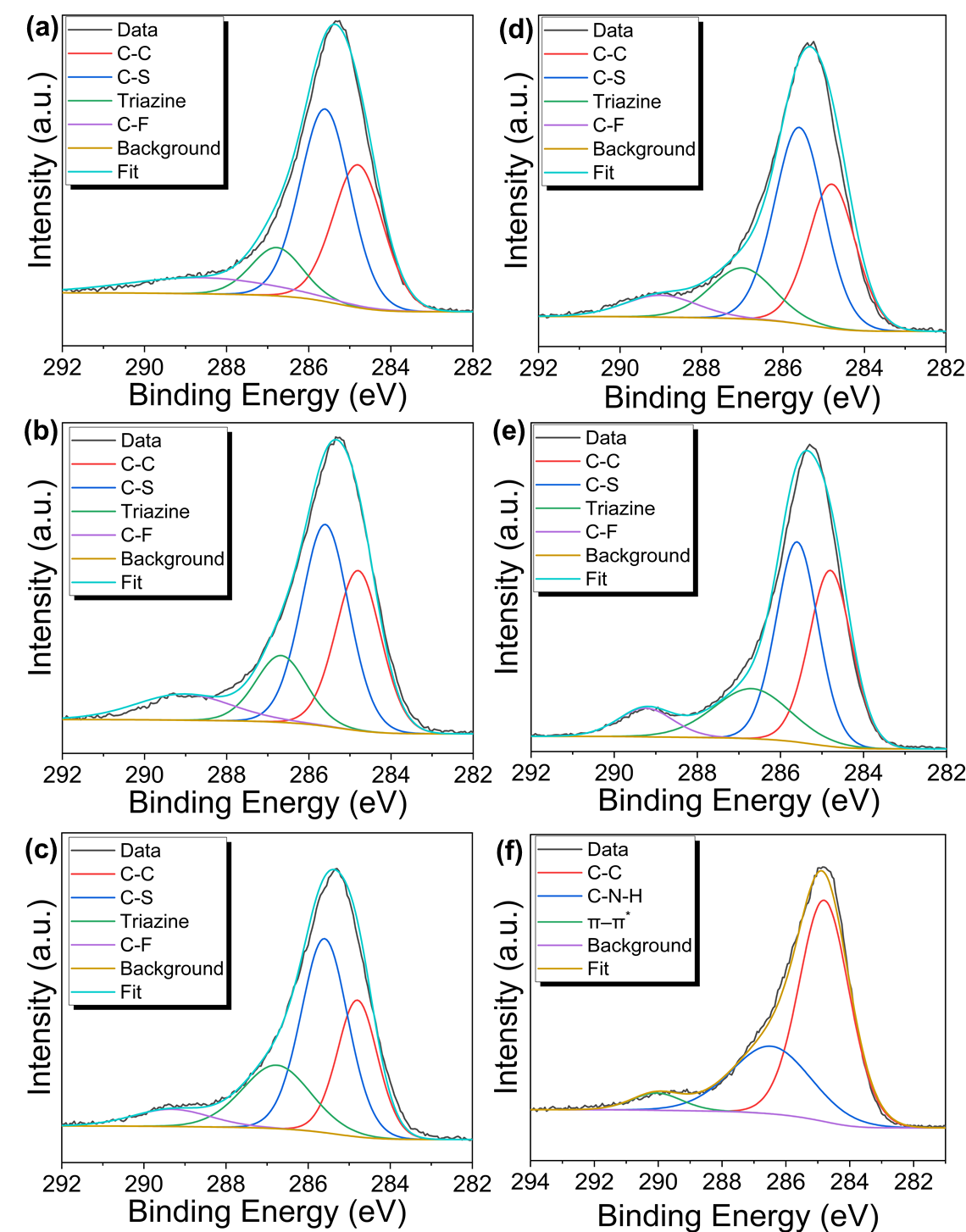
†Department of Materials Science and Engineering, Korea Advanced Institute of Science and Technology, 291 Daehak-ro, Yuseong-gu, Daejeon 34141, Republic of Korea

§Department of Chemistry, University of Fribourg, Chemin de Musee 9, Fribourg 1700, Switzerland.

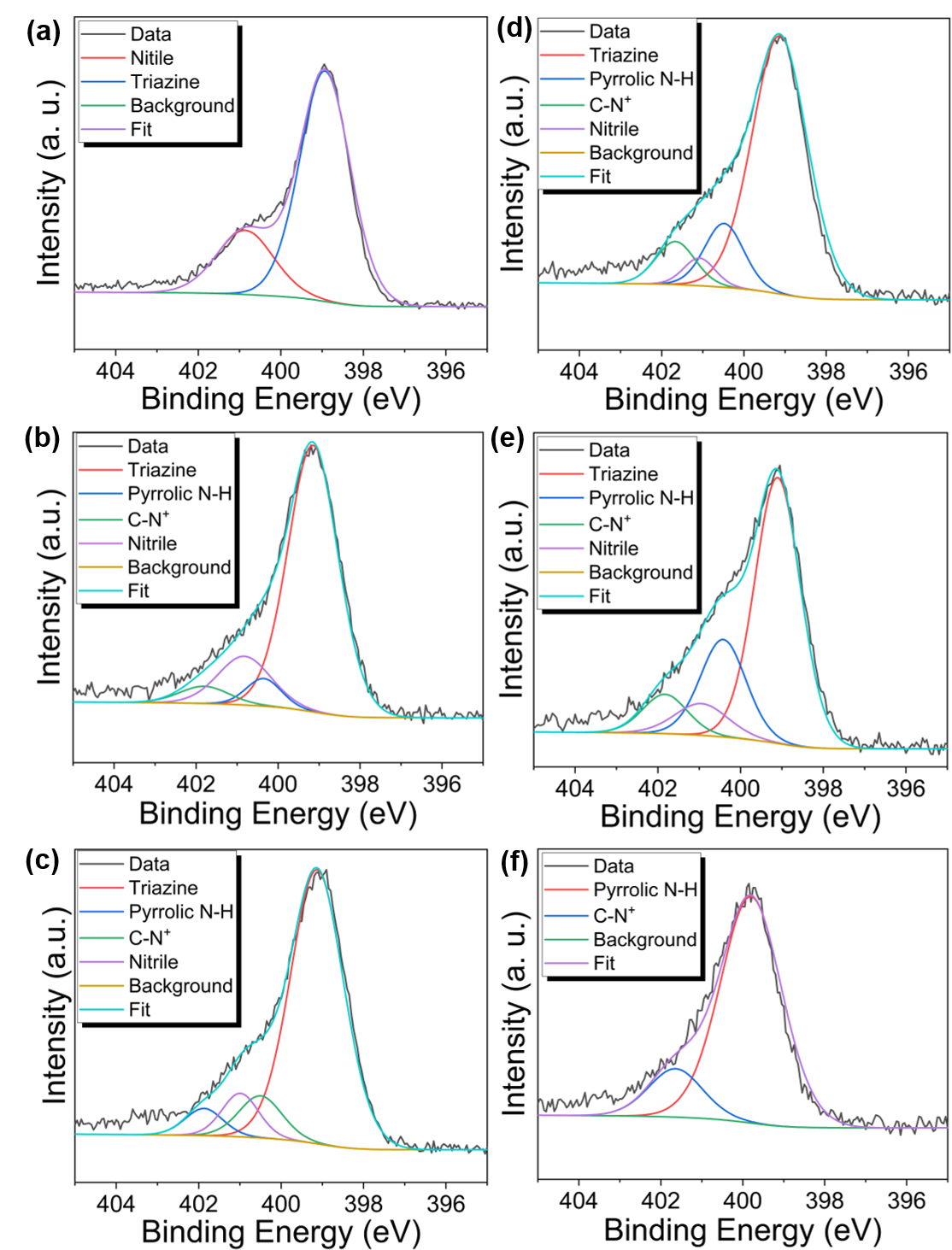
ҩSchool of Chemical and Biological Engineering and Institute of Chemical Processes, Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul 08826, Republic of Korea



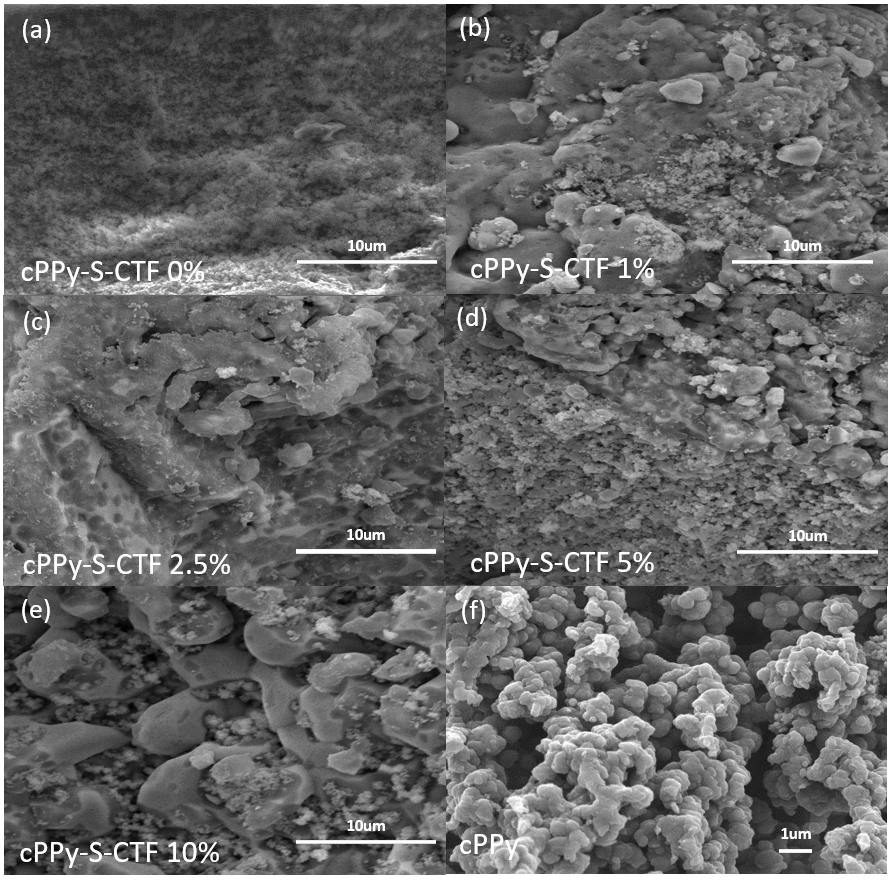
**Figure S1.** XRD spectra of elemental sulfur, cPpy-S-CTF 0%, cPpy-S-CTF 1%, cPpy-S-CTF 2.5%, cPpy-S-CTF 5%, and cPpy-S-CTF 10%.



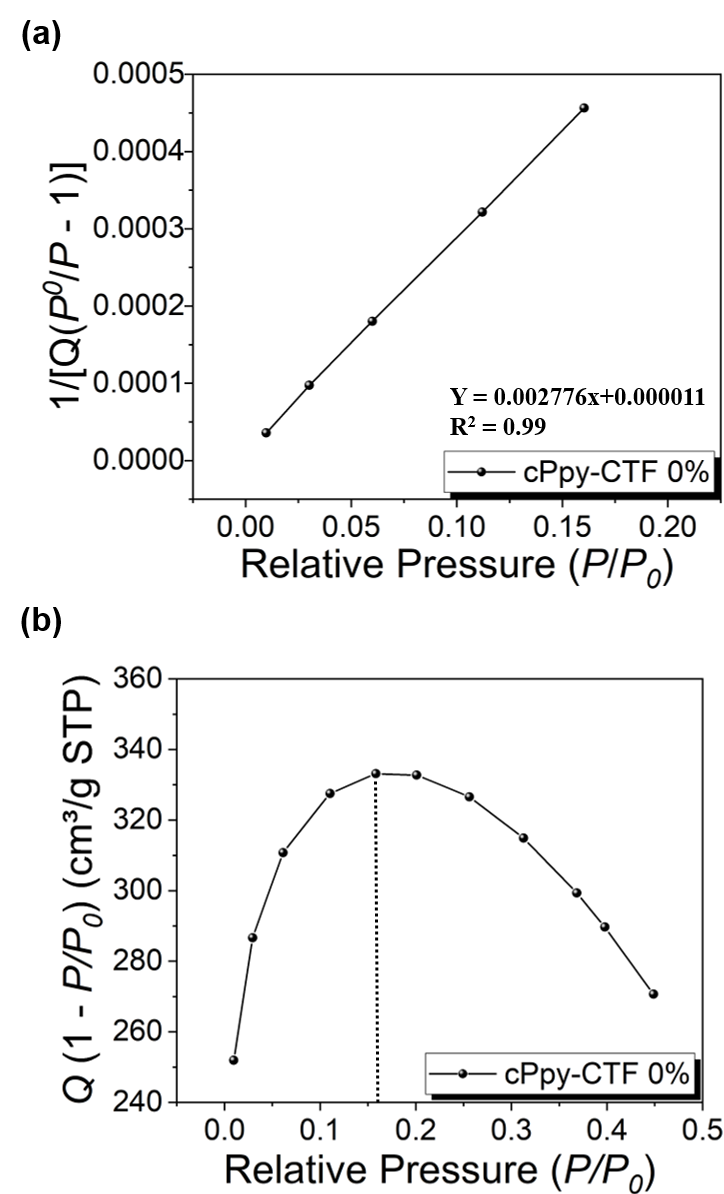
**Figure S2. C** 1s XPS profiles of (a) cPpy-S-CTF 0%, (b) cPpy-S-CTF 1% (c) cPpy-S-CTF 2.5%, (d) cPpy-S-CTF 5%, (e) cPpy-S-CTF 10%, and (f) cPpy.



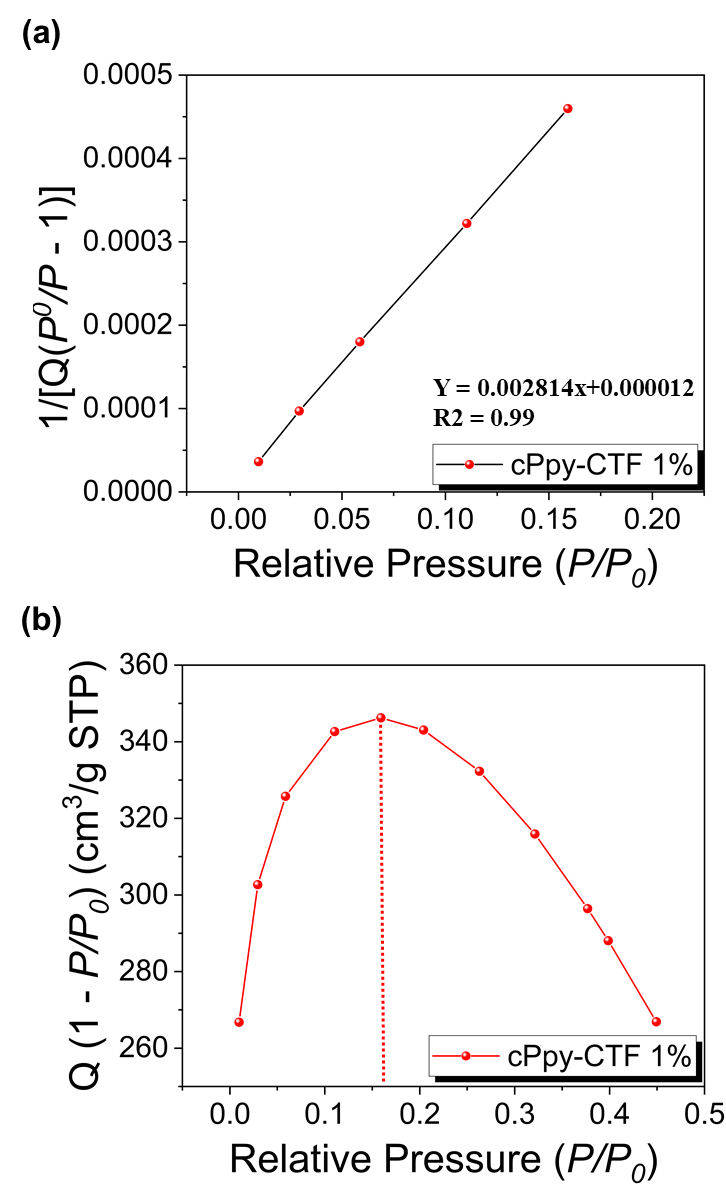
**Figure S3.** N 1s XPS profiles of (a) cPpy-S-CTF 0%, (b) cPpy-S-CTF 1% (c) cPpy-S-CTF 2.5%, (d) cPpy-S-CTF 5%, (e) cPpy-S-CTF 10%, and (f) cPpy.



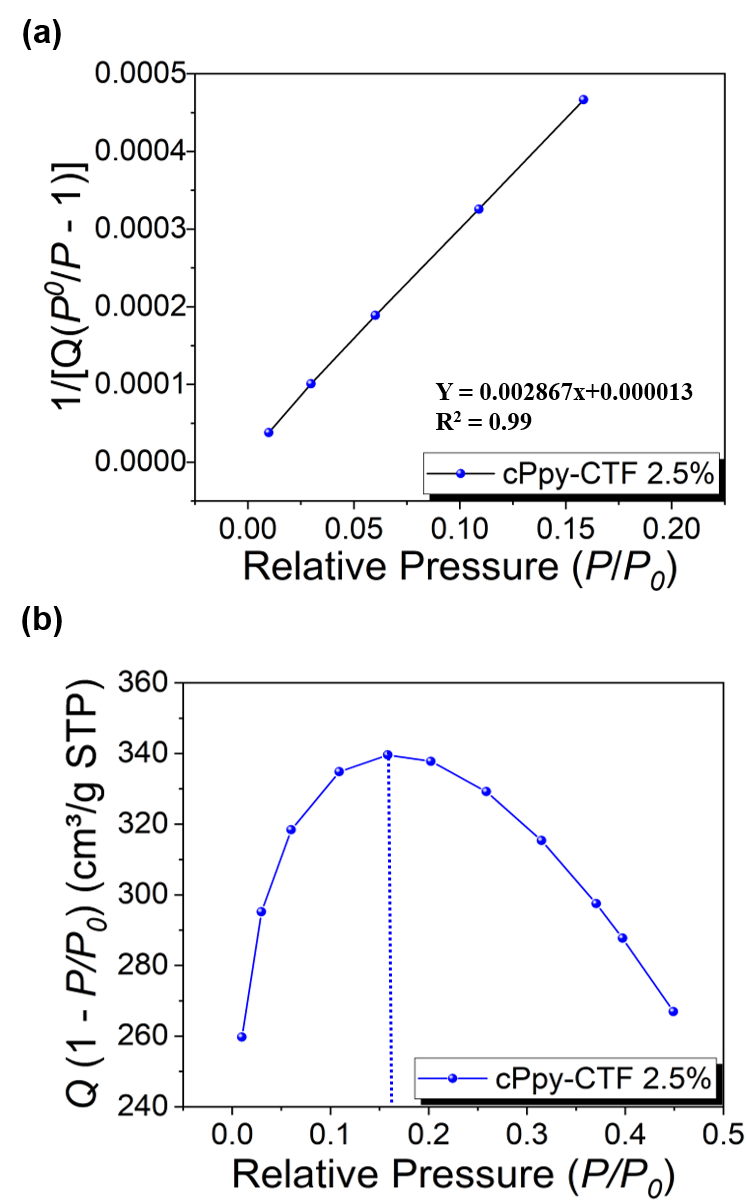
**Figure S4.** SEM images of various cPpy-S-CTFs. (a) cPpy-S-CTF 0%. (b) cPpy-S-CTF 1%. (c) cPpy-S-CTF 2.5%. (d) cPpy-S-CTF 5%. (e) cPpy-S-CTF 10%. (f) cPpy only.



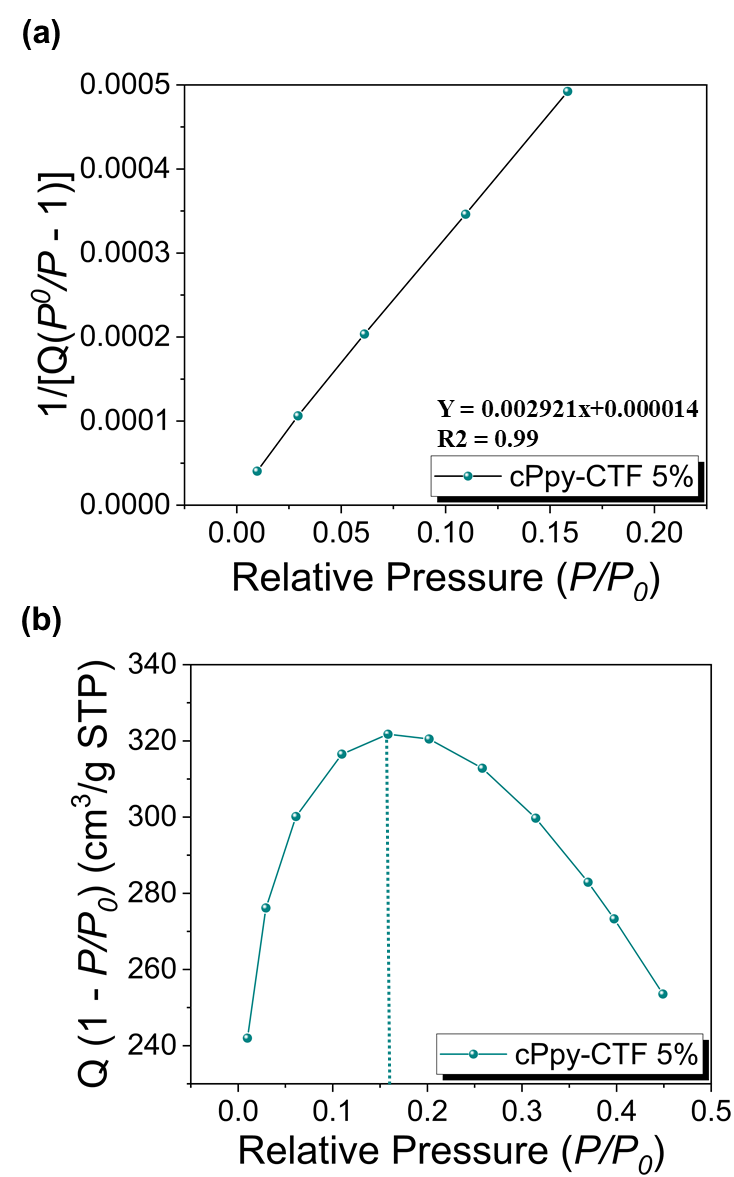
**Figure S5**. (a) Calculated Rouquerol plot of cPpy-CTF 0% according to pressure range used for the BET surface area calculation. (b) BET Linear plot of cPpy-CTF 0% obtained from nitrogen isotherms at 77 K. The selected points are located in the pressure ranges of 0.01 to 0.18 according to the Rouquerol plot.

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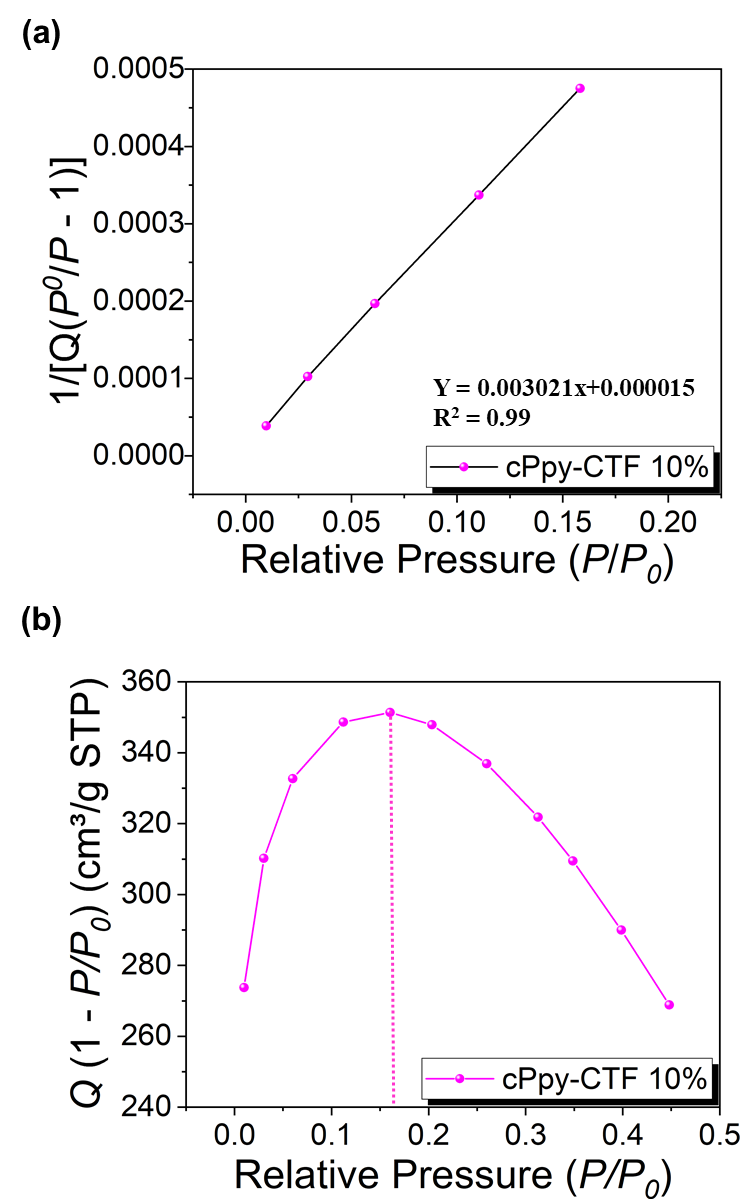
**Figure S6**. (a) Calculated Rouquerol plot of cPpy-CTF 1% according to pressure range used for the BET surface area calculation. (b) BET Linear plot of cPpy-CTF 1% obtained from nitrogen isotherms at 77 K. The selected points are located in the pressure ranges of 0.01 to 0.18 according to the Rouquerol plot.



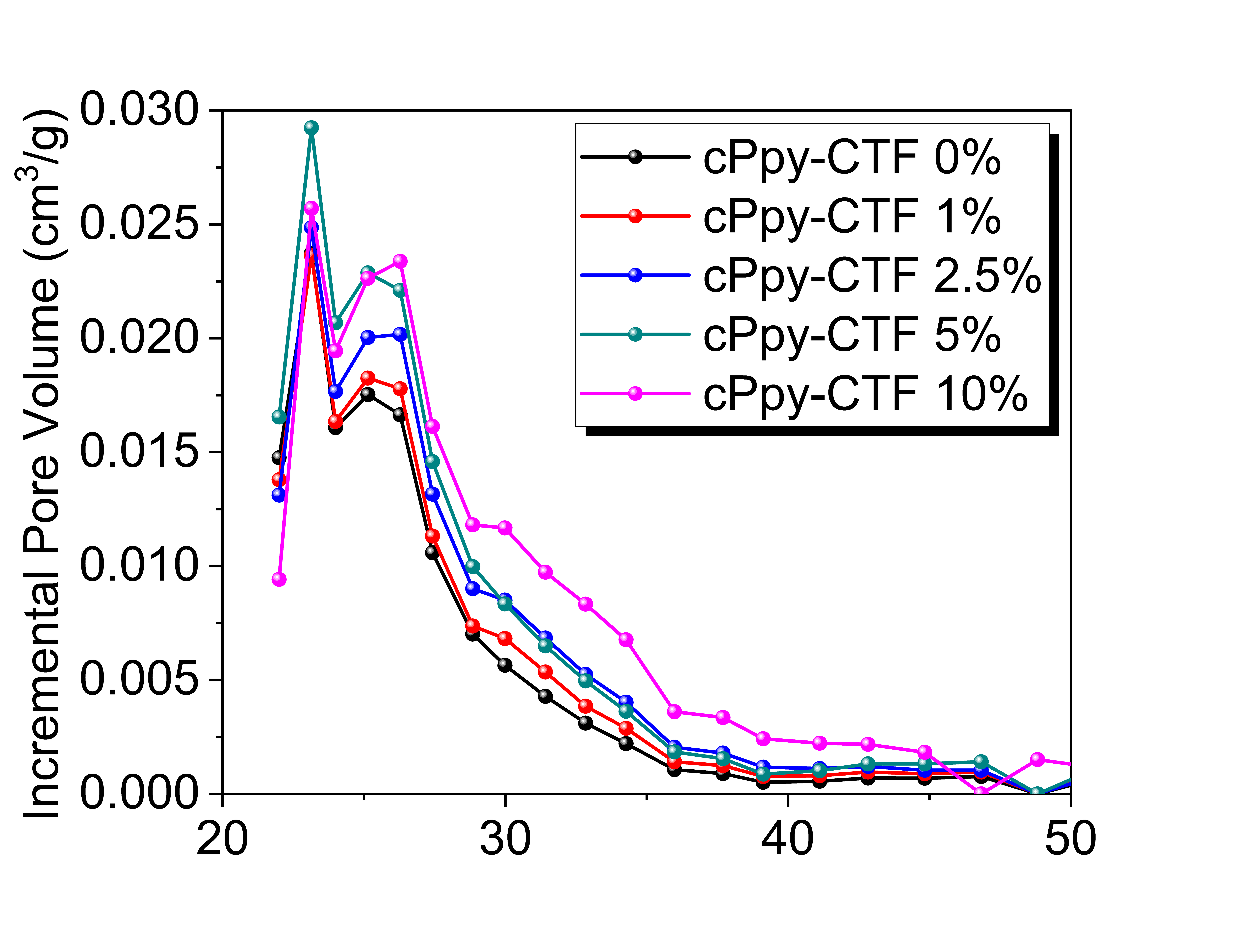
**Figure S7**. (a) Calculated Rouquerol plot of cPpy-CTF 2.5% according to pressure ra-nge used for the BET surface area calculation. (b) BET Linear plot of cPpy-CTF 2.5% obtained from nitrogen isotherms at 77 K. The selected points are located in the pressure ranges of 0.01 to 0.18 according to the Rouquerol plot.

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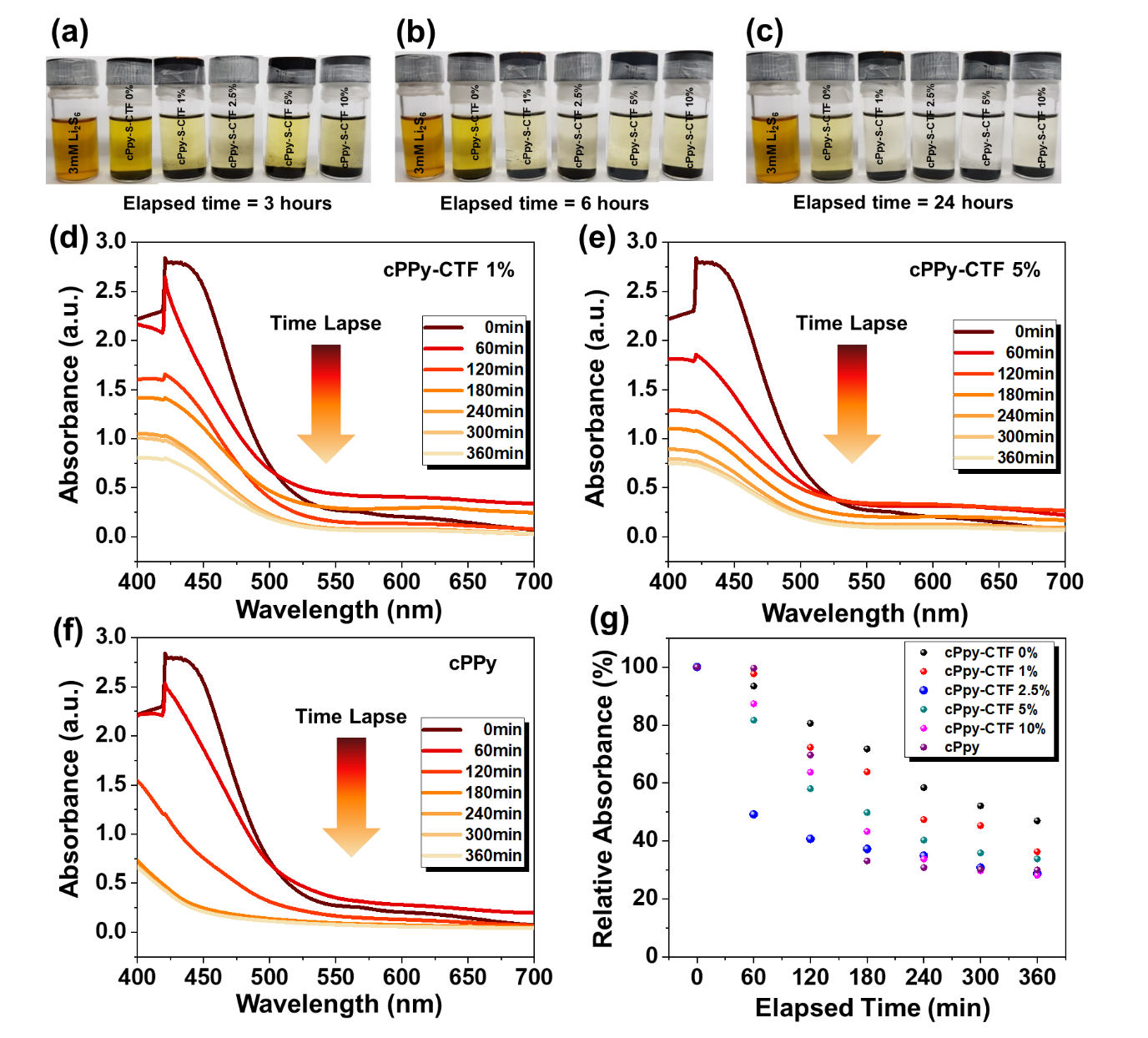
**Figure S8**. (a) Calculated Rouquerol plot of cPpy-CTF 5% according to pressure rang-e used for the BET surface area calculation. (b) BET Linear plot of cPpy-CTF 5% o-btained from nitrogen isotherms at 77 K. The selected points are located in the pressure ranges of 0.01 to 0.18 according to the Rouquerol plot.

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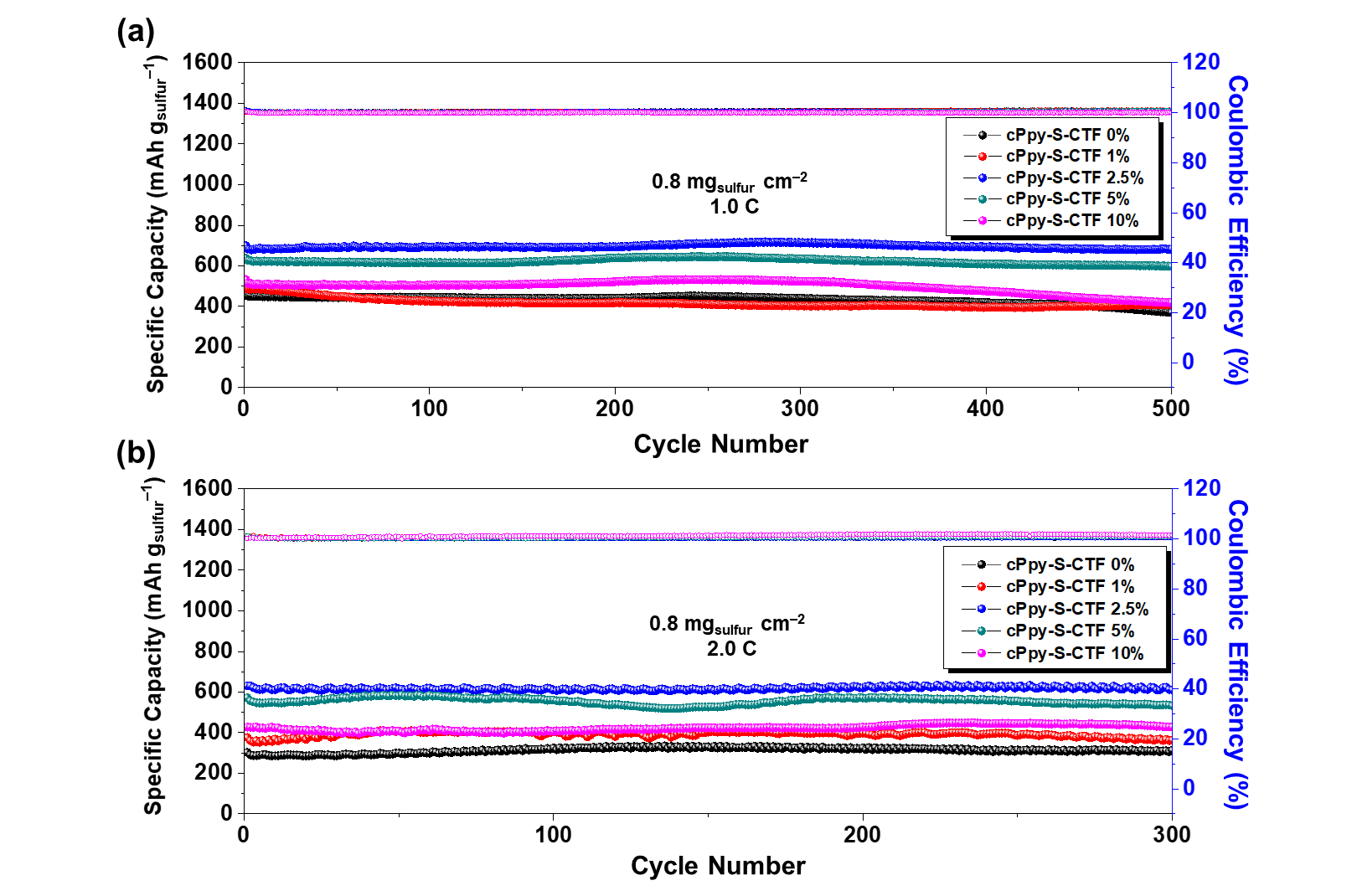
**Figure S9**. (a) Calculated Rouquerol plot of cPpy-CTF 10% according to pressure range used for the BET surface area calculation. (b) BET Linear plot of cPpy-CTF 10%obtained from nitrogen isotherms at 77 K. The selected points are located in the pre-ssure range of 0.01 to 0.18 according to the Rouquerol plot.



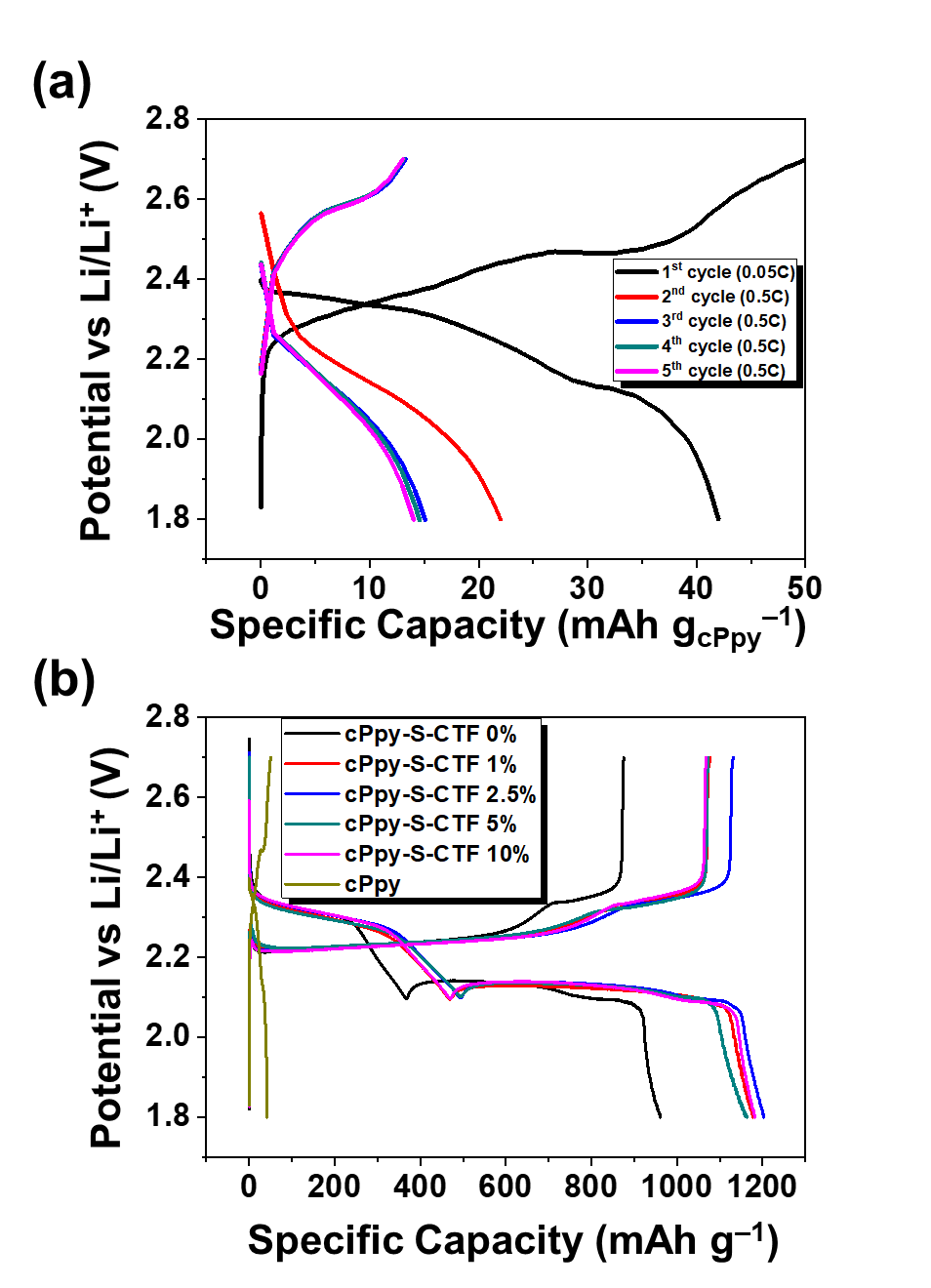
**Figure S10**. NLDFT pore size distributions of various cPpy-CTFs using standard carbon slit model.

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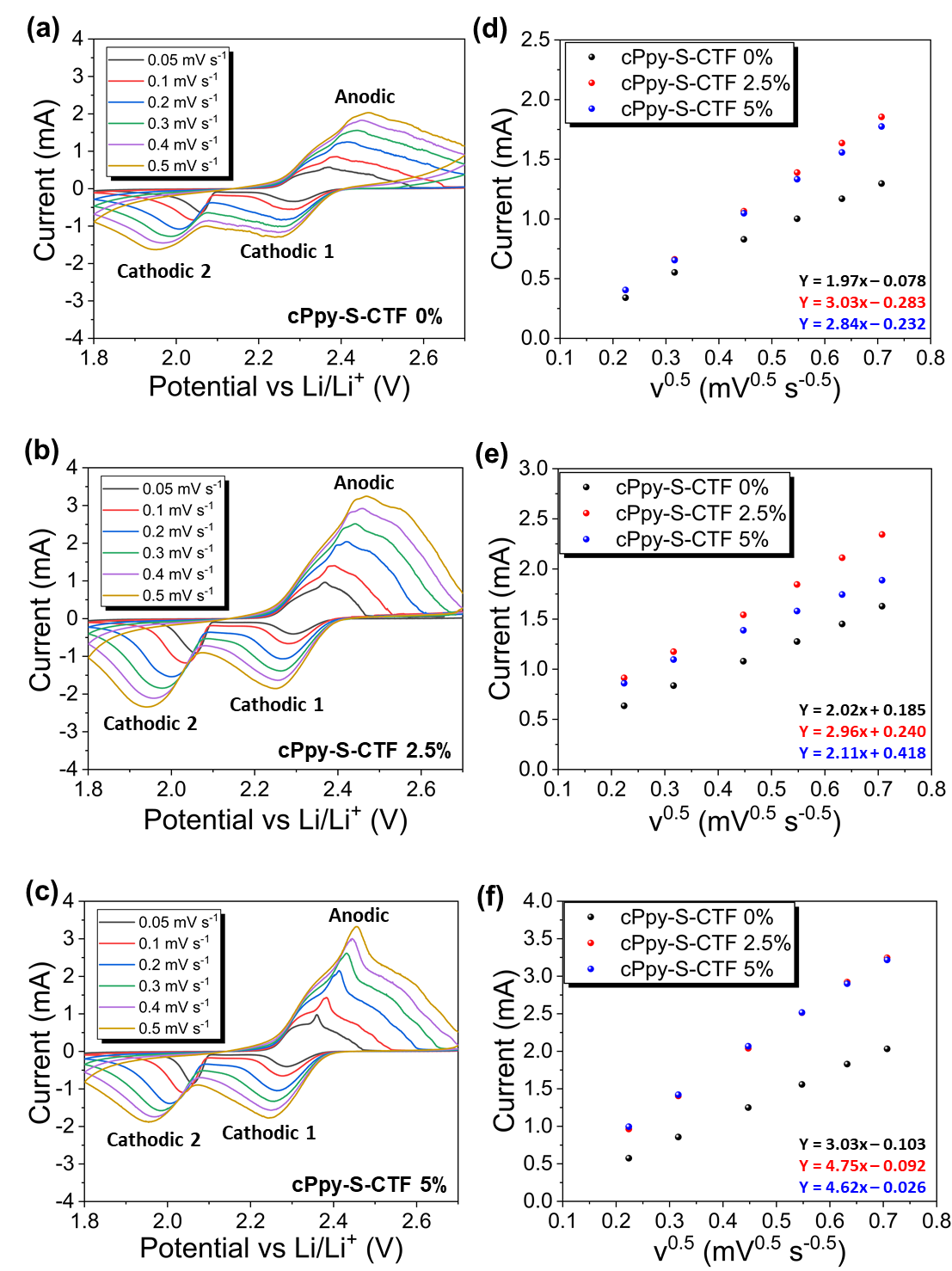
**Figure S11**. Visual effects of the 3 mM Li2S6 in DOL/DME (1:1 v/v) solution when exposed to various cPpy-CTFs over 3, 6 and 24 h (a, b, c). Time dependent UV-Vis absorption spectra of (d) cPpy-CTF 1%, (e) cPpy-CTF 5%, and (f) cPpy alone in 3 mM Li2S6 in DOL/DME (1:1 v/v) solution. (g) Plots of UV–Vis absorbance changes with time.



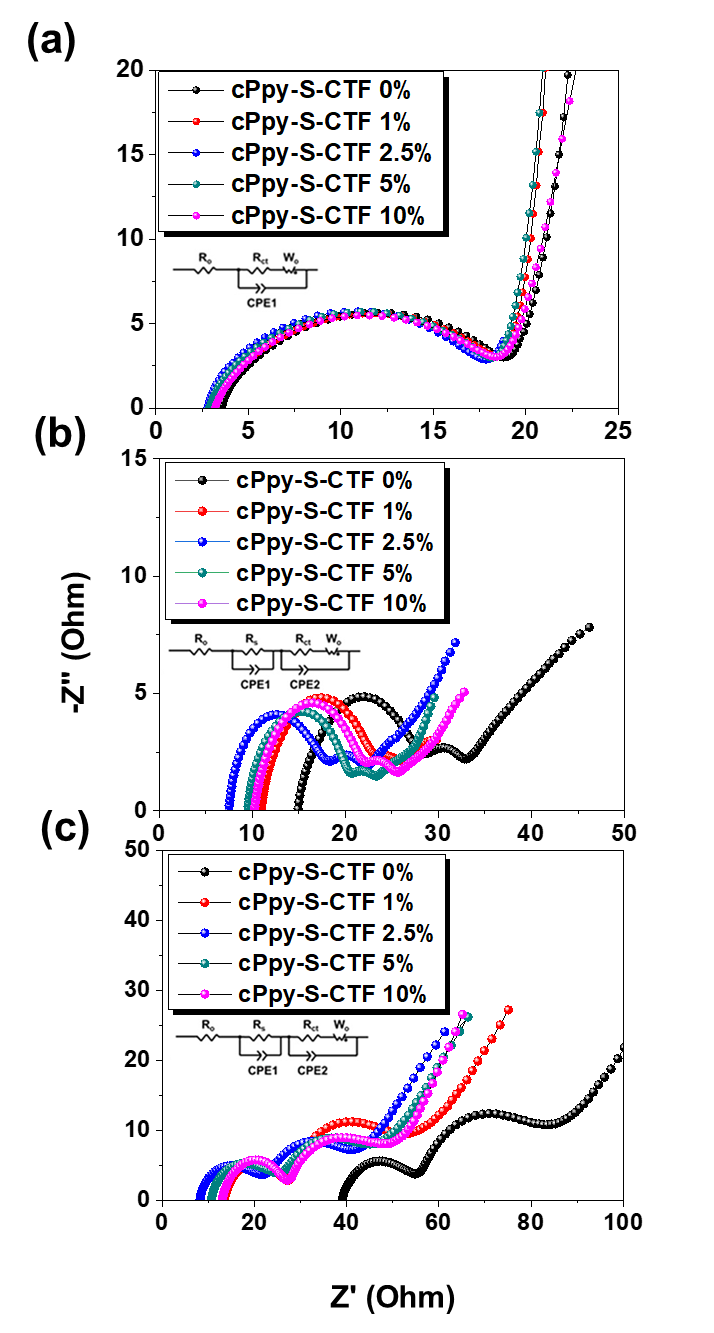
**Figure S12**. (a) Cycling performance and coulombic efficiencies of cPpy-S-CTFs at 1C and 0.8 mgsulfur cm–2 for 500 cycles. (d) Cycling performance and Coulombic efficiencies of cPpy-S-CTFs at 2C and 0.8 mgsulfur cm–2 for 300 cycles.

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**Figure S13**. (a) First discharge–charge profiles of bare cPpy at 0.05C (1st cycle) and 0.5C (subsequent cycles) in the potential range 1.8–2.7 V. (b) Overlaid first discharge-charge profiles of cPpy-S-CTFs and cPpy at 0.05C in the potential range 1.8–2.7 V.



**Figure S14.** CV plots of (a) cPpy-S-CTF 0%, (b) cPpy-S-CTF 2.5%, and (c) cPpy-S-CTF 5% in the potential range 1.8–2.7 V at various scan rates (0.05–0.5 mV s–1). Plots of CV peak current *vs*. the square root of scan rate for (d) cathodic reaction 1 (S8 → Li2S4), (e) cathodic reaction 2 (Li2S4 → Li2S), and (f) anodic reaction.



**Figure S15**. Nyquist plots of cPpy-S-CTFs (0–10%). (a) Before cycling, (b) after 100th charge, and (c) after 100th discharge.

**Table S1.** *ID*/*IG* values calculated from Raman spectroscopy of various cPpy-S-CTFs.

|  |  |  |  |
| --- | --- | --- | --- |
| **Samples** | ***ID* [a.u.]** | ***IG* [a.u.]** | ***ID*/*IG*** |
| cPpy-S-CTF 0% | 7897 | 8395 | 0.941 |
| cPpy-S-CTF 1% | 7858 | 8366 | 0.939 |
| cPpy-S-CTF 2.5% | 7651.5 | 7996 | 0.956 |
| cPpy-S-CTF 5% | 6986 | 7368 | 0.948 |
| cPpy-S-CTF 10% | 6738 | 7196 | 0.936 |

**Table S2.** Textural Properties ofvarious cPpy-S-CTFs.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Samples** | **BET a) [m2g–1]** | ***Smicro* b) [m2g–1]** | ***Sext*[m2g–1]** | ***Vmicro*c) [cm3g–1]** | ***Vext*d) [cm3g–1]** | ***Vtotal* e) [cm3g–1]** |
| CTF-0% Ppy | 1561 | 1472 | 89 | 0.70 | 0.10 | 0.80 |
| CTF-1% Ppy | 1539 | 1412 | 127 | 0.68 | .12 | 0.80 |
| CTF-2.5% Ppy | 1511 | 1379 | 132 | 0.67 | 0.13 | 0.80 |
| CTF-5% Ppy | 1497 | 1332 | 165 | 0.65 | .17 | 0.82 |
| CTF-10% Ppy | 1483 | 1298 | 184 | 0.64 | 0.19 | 0.83 |

a) Brunauer–Emmett–Teller (BET) surface area calculated over the pressure range (*P/Po*) of 0.01–1.0. b) Micro pore surface area calculated using *t*-plot method. c) Micro pore volume calculated using *t*-plot method. Total pore volume obtained at *P*/*Po* = 0.99. e) *Vext* =*Vtotal* – *Vmicro*.

**Table S3.** EIS results of cPpy-S-CTF 0, 1, 2.5, 5, and 10% before cycling, after 100 cycles of full charge and discharge.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Samples** | **Resistance** | **Before Cycling** | **After 100th cycle**  **(Full charge)** | **After 100th cycle**  **(Full discharge)** |
| cPpy-S-CTF 0% | *Ro*[Ω] | 3.5 | 15.6 | 40.3 |
| *Rct*[Ω] | 16.7 | 6.0 | 21.5 |
| *Rs*[Ω] | - | 11.2 | 14.0 |
| cPpy-S-CTF 1% | *Ro*[Ω] | 3.1 | 10.9 | 13.4 |
| *Rct*[Ω] | 16.4 | 5.8 | 20.2 |
| *Rs*[Ω] | - | 11.8 | 12.8 |
| cPpy-S-CTF 2.5% | *Ro*[Ω] | 2.7 | 7.8 | 8.7 |
| *Rct*[Ω] | 15.7 | 5.1 | 15.2 |
| *Rs*[Ω] | - | 9.1 | 11.8 |
| cPpy-S-CTF 5% | *Ro*[Ω] | 2.8 | 9.5 | 10.6 |
| *Rct*[Ω] | 15.4 | 4.6 | 15.9 |
| *Rs*[Ω] |  | 10.1 | 12.4 |
| cPpy-S-CTF 10% | *Ro*[Ω] | 3.2 | 10.1 | 13.0 |
| *Rct*[Ω] | 14.7 | 5.2 | 18.9 |
| *Rs*[Ω] |  | 10.9 | 13.9 |