

## Supporting Information

### Enhanced Dielectric Performance with High-Temperature Stability by Interface- Modulation of the Core-Shell Structured Imide-Polymer@BT Nanohybrids in PEI-Based Nanocomposites

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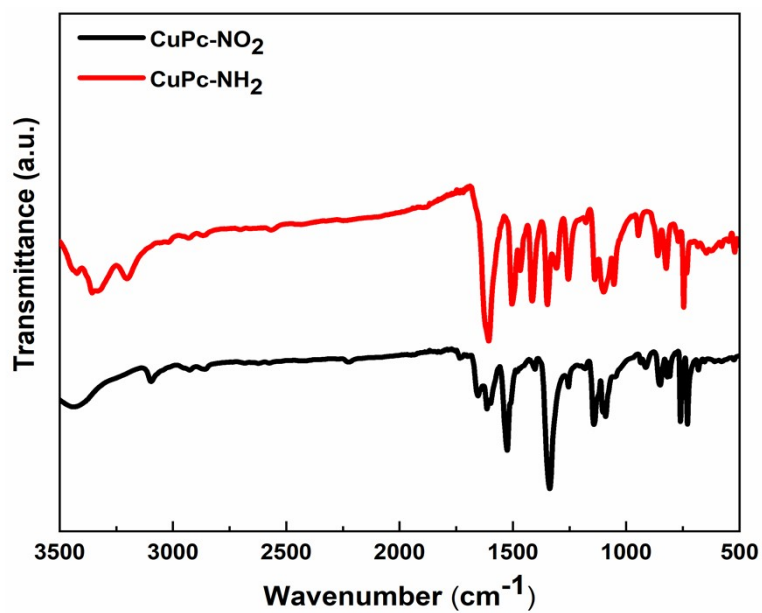


Fig. S1 FTIR spectra of CuPc-NO<sub>2</sub> and CuPc-NH<sub>2</sub>

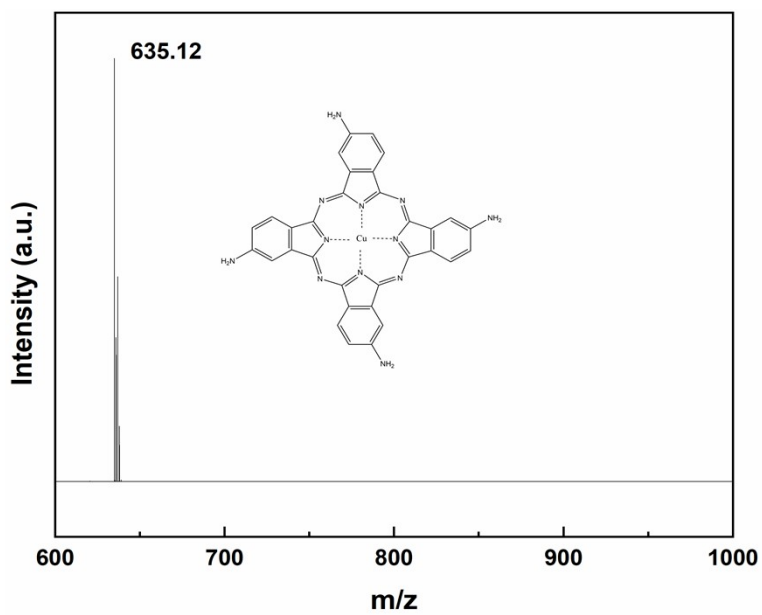
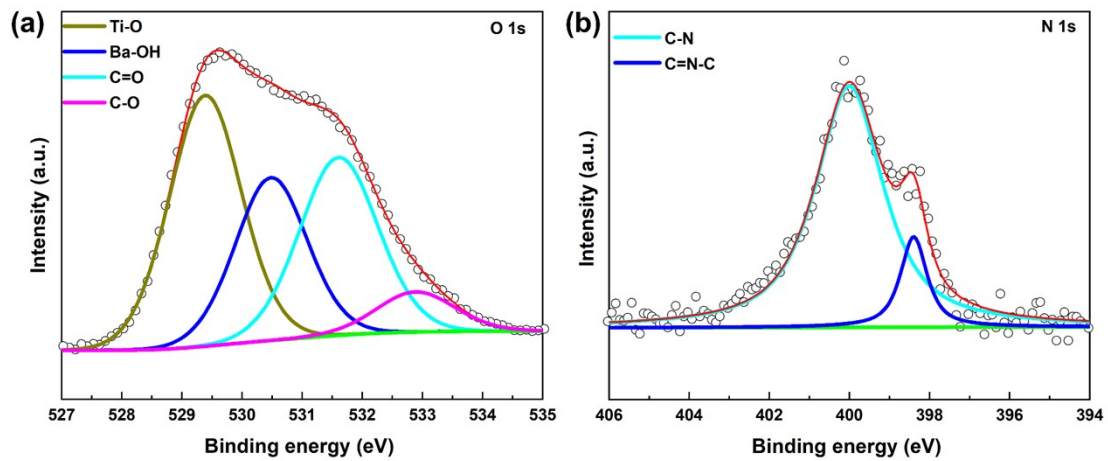
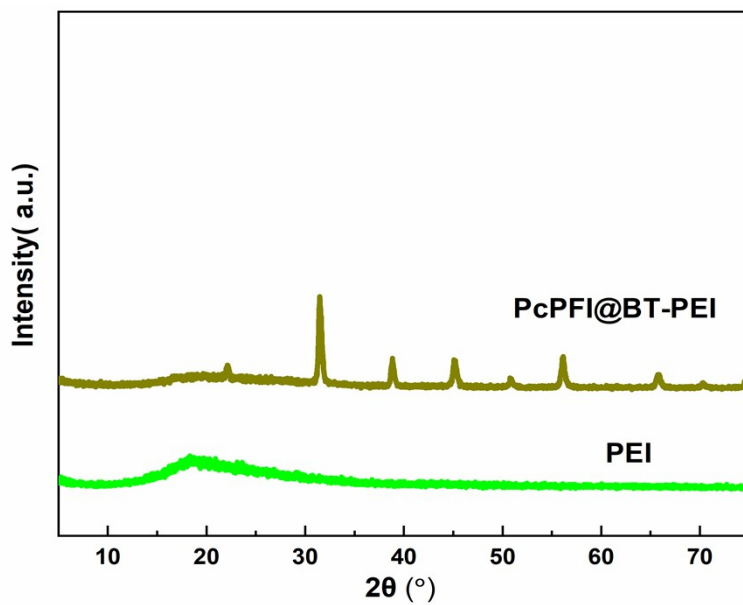


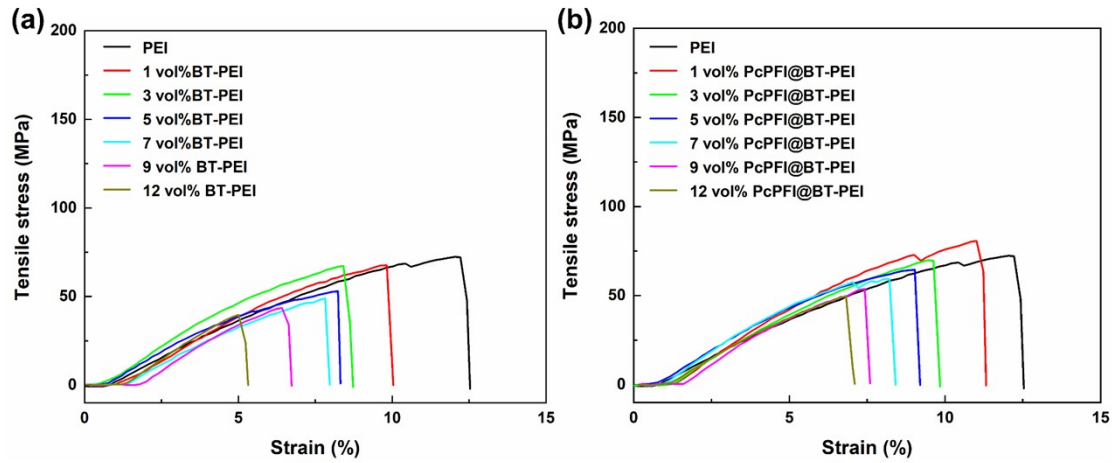
Fig. S2 Mass spectrometry of CuPc-NH<sub>2</sub>



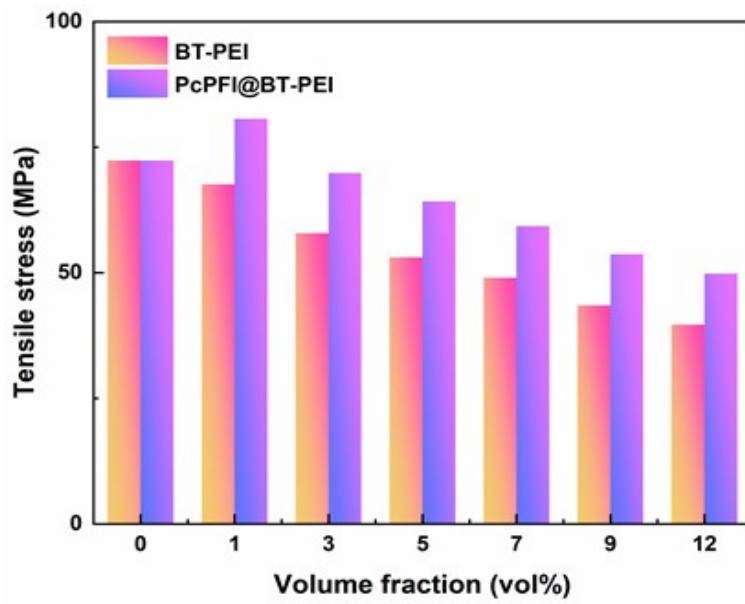
**Fig. S3** High-resolution O 1s (a) and N 1s (b) spectra of PcPFI@BT



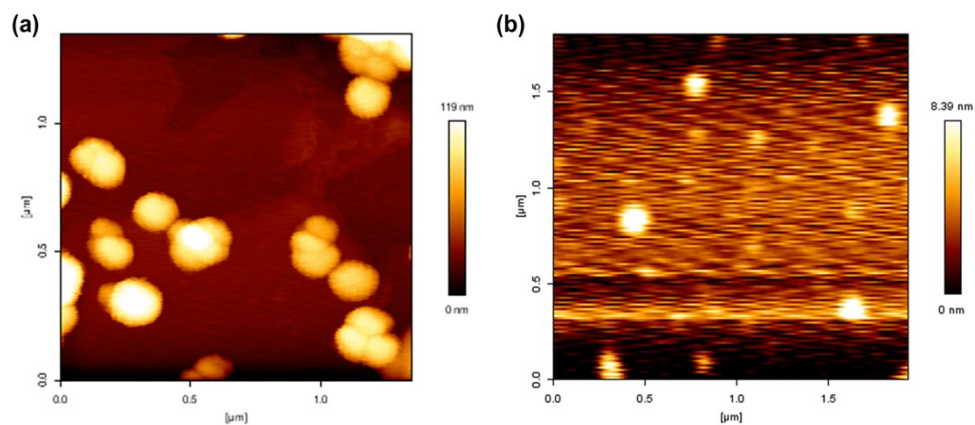
**Fig. S4** XRD patterns of PEI and 5vol% PcPFI@BT-PEI



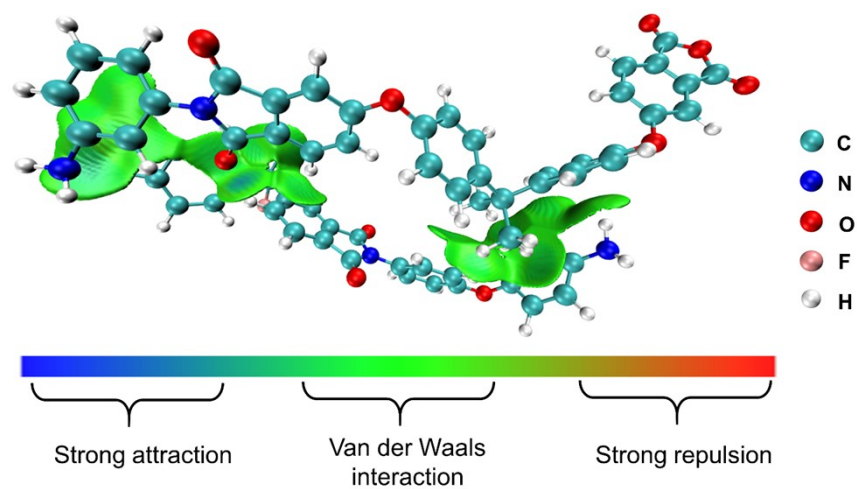
**Fig. S5** Stress-strain curves of BT-PEI (a) and PcPFI@BT-PEI (b) with the filler content from 0 to 12 vol%



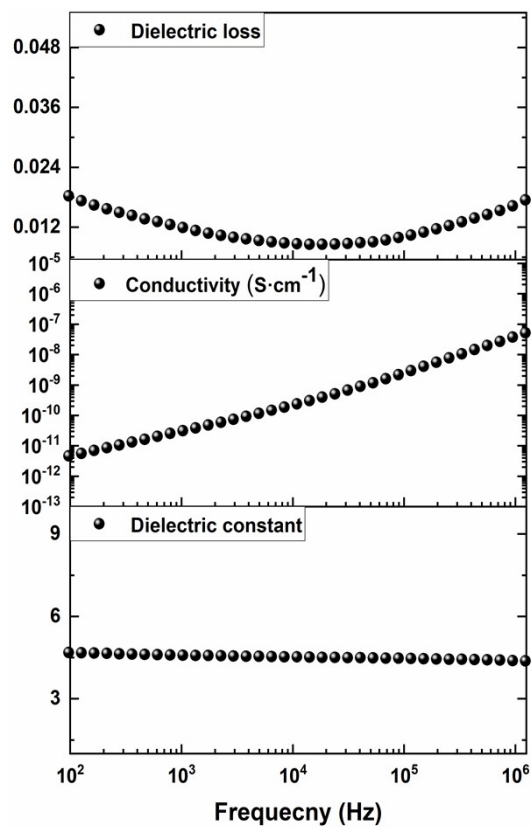
**Fig. S6** Tensile stress of the two sets of composites with filler content from 0 to 12 vol%



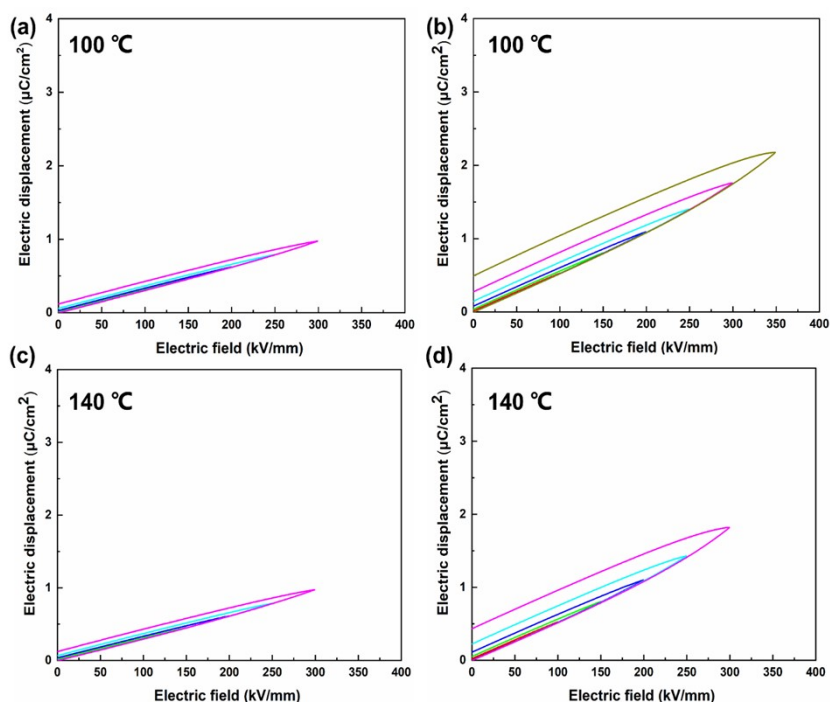
**Fig. S7** AFM images of BT-PEI (a) and PcPFI@BT-PEI (b)



**Fig. S8** The intermolecular interaction between the similar components of PEI and PcPFI



**Fig. S9** Dielectric properties of PcPFI polymer from  $10^2$  to  $10^6$  Hz



**Fig. S10** D-E loops of PEI ((a), (c)) and 1 vol% PcPFI@BT-PEI ((b), (d)) at 100 °C and 140 °C

**Table S1 The dielectric properties of the prepared composite and some related polymeric dielectrics reported in previous reports**

| Materials  | Dielectric Constant (1 kHz) | Dielectric Loss (1 kHz) | Breakdown Strength (kV/mm) | Energy storage density (J/cm <sup>3</sup> ) | Charge-discharge efficiency (%) | Ref              |
|--|-----------------------------|-------------------------|----------------------------|---|---------------------------------|------------------|
| 10 vol% TiO <sub>2</sub> -BT-TiO <sub>2</sub> @dopa/PVDF                     | 12.6                        | 0.05                    | 312.8                      | 4.4   | ~30                             | S1               |
| 2 wt% PDA@BT/c-PVDF  | ~11                         | ~0.07                   | 242                        | 2.9   | 75                              | S2               |
| 20 vol% BT-HCuPc/PES   | ~9.8                        | ~0.044                  | ~290                       | 2.0   | 84                              | S3               |
| 5 wt% BT@PI/PVDF   | 15                          | 0.046                   | 178.2                      | Not given                                   | Not given                       | S4               |
| 3 vol% BT-Bi(Li <sub>0.5</sub> Nb <sub>0.5</sub> )O <sub>3</sub> /P(VDF-HFP) | ~10                         | ~0.025                  | 497                        | 14.2  | 55.5                            | S5               |
| 10 wt% BNNSs/RC  | 7                           | 0.02                    | 370                        | 4.1   | ~75                             | S6               |
| 10 vol% BT-1.2 vol% GO/P(VDF-HFP)  | 66.2                        | 0.048                   | 40                         | Not given                                   | Not given                       | S7               |
| 1 wt% HfO <sub>2</sub> @BT/P(VDF-HFP)  | ~10                         | ~0.016                  | 466.1                      | 10.7  | 68.1                            | S8               |
| 10 wt% BT-Ni(OH) <sub>2</sub> /P(VDF-HFP)                                    | 14.8                        | 0.038                   | 330                        | 7.13<br>(calculated)                        | Not given                       | S9               |
| <b>1 vol% PcPFI@BT/PEI</b>   | <b>4.35</b>                 | <b>0.014</b>            | <b>381</b>                 | <b>3.04</b>                                 | <b>89.25</b>                    | <b>This work</b> |

## Reference

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