Largely Enhanced Dielectric Constant of PVDF Nanocomposites through a Core-Shell Strategy

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Fig. S1 XRD patterns of TiO$_2$@C NWs hybrids with different carbon shell thickness.

Fig. S2 Raman spectra of TiO$_2$@C NWs hybrids with different carbon shell thickness.
Fig. S3 FT-IR spectra of (a) PVDF and TiO$_2$@C-15 NWs/PVDF nanocomposites with different filler loadings and (b) TiO$_2$ NWs/PVDF and TiO$_2$@C NWs/PVDF nanocomposites with different hybrids at the same filler loading (15 vol. %); (c) The calculated relative content of $\alpha$ and $\beta$ phase in PVDF and TiO$_2$@C-15 NWs/PVDF nanocomposites as a function of filler loading; (d) The calculated relative content of $\alpha$ and $\beta$ phase in TiO$_2$ NWs/PVDF and TiO$_2$@C NWs/PVDF nanocomposites at 15 vol. % filler loading as a function of CVD time.
Fig. S4 XRD patterns of (a) PVDF and TiO$_2$@C-15 NWs/PVDF nanocomposites with different filler loadings and (b) TiO$_2$ NWs/PVDF and TiO$_2$@C NWs/PVDF nanocomposites with different hybrids at the same filler loading (15 vol. %); (c) The calculated content of $\beta$ phase in total crystalline phase of PVDF and TiO$_2$@C-15 NWs/PVDF nanocomposites as a function of filler loading; (d) The calculated content of $\beta$ phase in total crystalline phase of TiO$_2$ NWs/PVDF and TiO$_2$@C NWs/PVDF nanocomposites at 15 vol. % filler loading as a function of CVD time.
Fig. S5 Origin’s multiple peak separation fitting results of PVDF and TiO$_2$@C-15 NWs/PVDF nanocomposites with different filler loadings.

Fig. S6 Origin’s multiple peak separation fitting results of TiO$_2$ NWs/PVDF and TiO$_2$@C NWs/PVDF nanocomposites with different hybrids at the same filler loading (15 vol. %).
Fig. S7 Frequency dependence of the dielectric loss values of TiO$_2$@C-5 NWs/PVDF, TiO$_2$@C-15 NWs/PVDF and TiO$_2$@C-45 NWs/PVDF nanocomposites at different filler loadings.

Fig. S8 Typical variation of dielectric loss values (10$^3$ Hz) of TiO$_2$ NWs/PVDF, TiO$_2$@C-5 NWs/PVDF, TiO$_2$@C-15 NWs/PVDF, and TiO$_2$@C-45 NWs/PVDF nanocomposites as a function of carbon shell thickness at different filler loadings (15 vol. % and 20 vol. %).
Fig. S9 Dependence of the dielectric loss and AC conductivity of TiO$_2$@C-5 NWs/PVDF, TiO$_2$@C-15 NWs/PVDF, and TiO$_2$@C-45 NWs/PVDF nanocomposites on the volume fraction of the TiO$_2$ NWs core and carbon shell for the whole nanocomposites ($10^3$ Hz).

Fig. S10 The D-E loops of (a) PVDF and TiO$_2$@C-15 NWs/PVDF nanocomposites with different filler loadings (b) TiO$_2$ NWs/PVDF and TiO$_2$@C NWs/PVDF nanocomposites with different hybrids at the same filler loading (5 vol. %) at 10 Hz.
Fig. S11 Comparison of dielectric properties ($10^3$ Hz) of percolative nanocomposites with different types of nano-fillers: TiO$_2$@C NWs, 2.5-20 vol. %; Flower-like TiO$_2$-C, 15 and 20 vol. %; BT@C, 5-30 vol. %; BT-Ag, 7.6 and 18 vol. %; PPy nanoclips, 3-9 wt. %; rGO-CNTs, 0.02-0.144 wt. % [1, 2, 3, 4, 5].

The derivation process of volume fraction of carbon shell ($\alpha$):

The volume fraction of carbon shell in the hybrids ($\alpha$) could be calculated from the weight fraction of carbon shell ($\beta$) as described below. The weight fraction of carbon shell in the hybrids could be obtained from the TGA curves. The equation for the calculation of $\alpha$ could be expressed as below.

$$\alpha = \frac{V_c}{V_h} = \frac{V_c}{V_c + V_{TiO2}} = \frac{\beta}{\beta M/\rho_c + (1 - \beta) M/\rho_{TiO2}} = \frac{\beta/\rho_c}{\beta/\rho_c + (1 - \beta)/\rho_{TiO2}}$$

where $V_c$, $V_{TiO2}$, and $V_h$ are the volume of carbon shell, TiO$_2$ core, and hybrids, respectively. The $\rho_c$ and $\rho_{TiO2}$ represent the density of carbon shell and TiO$_2$ core, respectively, and $M$ is the mass of hybrids. The $\rho_c$ and $\rho_{TiO2}$ values are selected as 2.00 g cm$^{-3}$ and 3.90 g cm$^{-3}$, respectively. The $\beta$ is directly extracted from the TGA results.
Then the corresponding numerical values are put into the above-mentioned equation, and then the corresponding $\alpha$ could be obtained. The $\alpha$ for the TiO$_2$@C-5 NWs, TiO$_2$@C-15 NWs, and TiO$_2$@C-45 NWs are 8.34 %, 27.99 %, and 40.95 %, respectively.


