Supporting Information for:

Highly self-healable and recyclable graphene nanocomposites composed of a Diels–Alder crosslinking/P3HT nanofibrils dual-network for electromagnetic interference shielding

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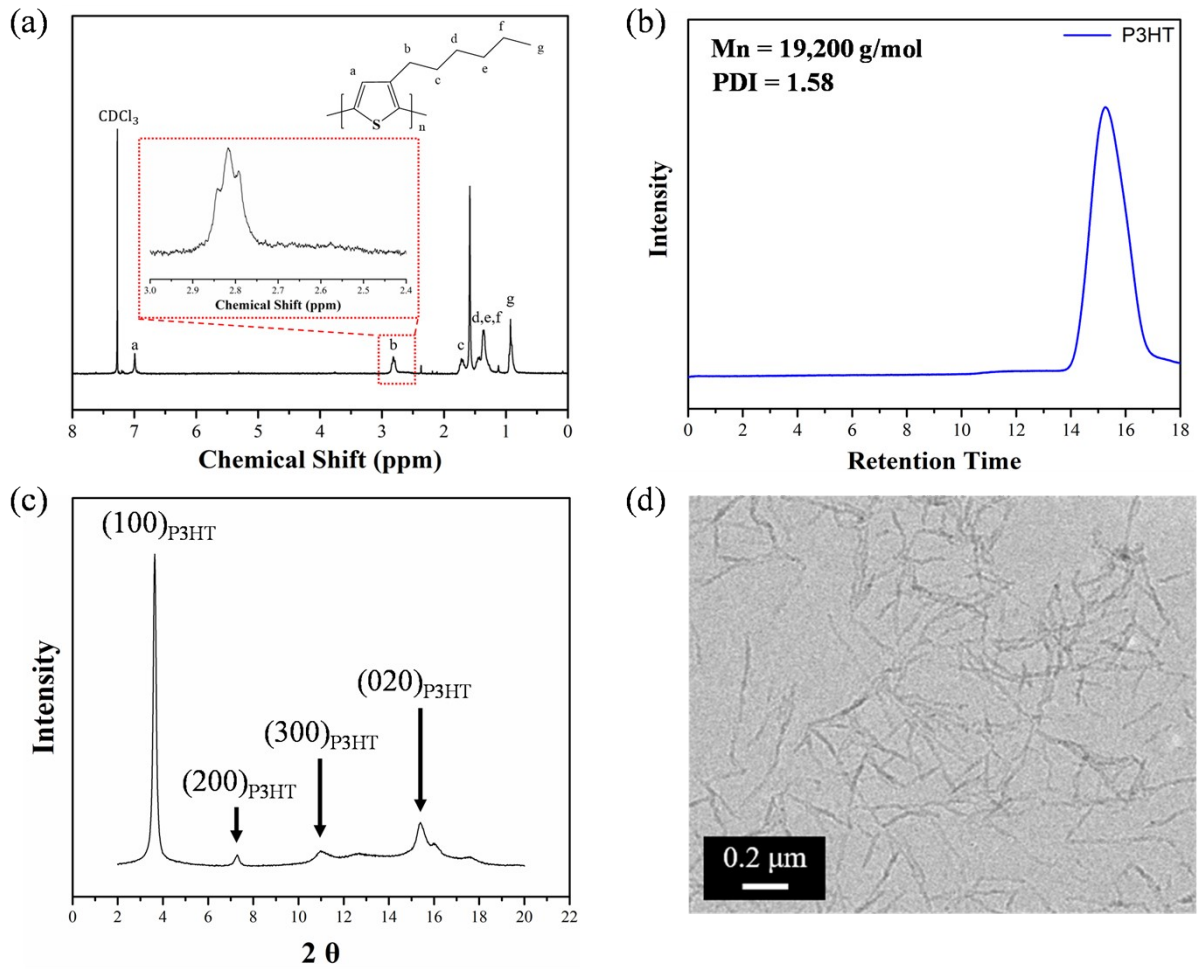

Figure. S1 (a) ^{1}H NMR spectra of the synthesized P3HT sample. The inset figure shows the magnified profile from 2.4 to 3.0 ppm. (b) GPC profile of the synthesized P3HT sample. (c)WAXS profile of the synthesized P3HT sample. (d) TEM micrograph of the synthesized P3HT sample which shows the formation of nanofibrils in a toluene/anisole mixed solvent (2:1 toluene-anisole volume ratio) after aging for 7 days.

The ^{1}H NMR spectrum of the synthesized P3HT is shown in Fig. S1. The characteristic signals of the ^{1}H NMR profile (Fig. S1a) effectively proved the successful synthesis of P3HT via the Grignard metathesis method. Additionally, the signals located at 2.82 ppm and 2.56 ppm were associated with the α-methylene protons of the hexyl side chain in head-to-tail and head-to-head configurations, respectively. Note that the head-to-head signal was nearly invisible, indicating that this sample possessed a high degree of configurational regularity. The molecular weight characteristics of the synthesized P3HT were further measured by GPC. The GPC profile of this P3HT sample revealed a single main peak, as shown in Fig. S1b. The number-average molecular weight (Mn) and polydispersity index were 19,200 g/mol and 1.58, respectively. The crystalline structure of the synthesized P3HT was further identified by wide-

angle X-ray scattering (WAXS) measurement. The WAXS experiment was carried out in 13A1 beamline of NSRRC in Taiwan. From the experiment result shown in Fig. S1c, it was found that the WAXS profile of the P3HT sample showed significant $(100)_{P3HT}$, $(200)_{P3HT}$, $(300)_{P3HT}$, and $(020)_{P3HT}$ crystalline peaks, indicating that this highly regioregular P3HT exhibited a good crystallization ability that could facilitate the molecular chains to organize into a highly ordered crystalline structure. Additionally, the excellent driving force for crystallization of this synthesized P3HT could also endow the synthesized P3HT with the ability of solution crystallization in a marginal solution environment, inducing gradual stacking of the P3HT molecular chains, leading to a formation of nanofibrils. Fig. S1d shows the self-assembled nanofiber structure of the synthesized P3HT in a toluene/anisole mixed solvent (2:1 toluene-anisole volume ratio) after aging for 7 days. Since anisole is a marginal solvent of P3HT, this result clearly confirmed that the highly regioregular P3HT in the current study possessed remarkable capability of solution crystallization and could be used to manufacture highly ordered nanofibers in the solution medium.

Table S1. Thermal stability of the PHBCF, PG1, PG2.5, PG2.5H2.5, and PG5H5 samples

Sample Code	**5wt% loss temperature (°C)** [1]
PHBCF	352.3
PG1	377.3
PG2.5	379.4
PG2.5H2.5	376.7
PG5H5	381.9

[1] 5wt% loss temperatures were identified from TGA thermograms.

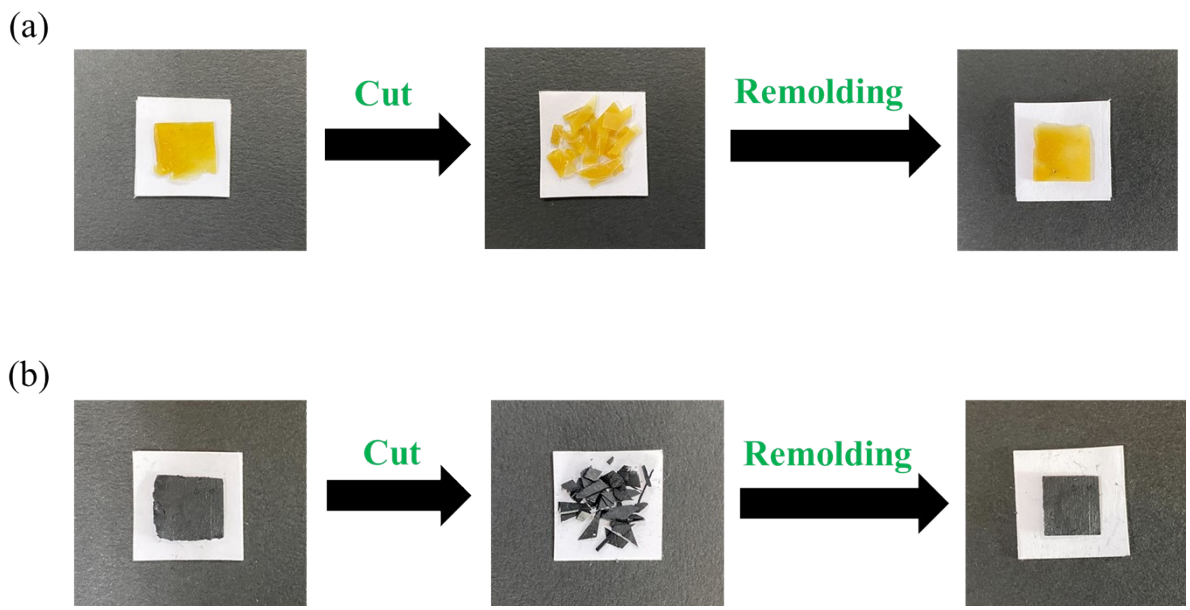

Figure S2. Thermal remolding process of the (a) PG0 and (b) PG2.5 nanocomposites.

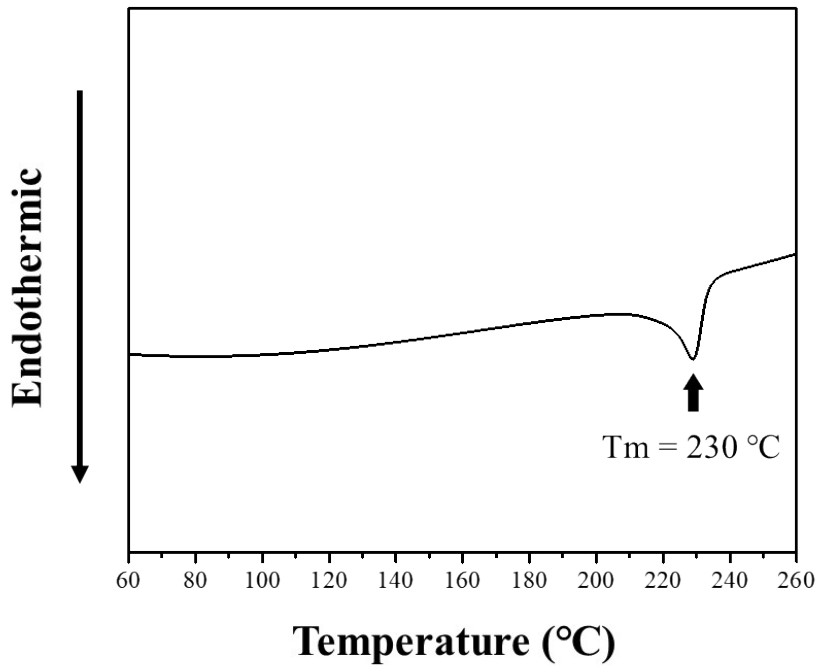

Figure S3. DSC heating curve of the pristine P3HT sample.

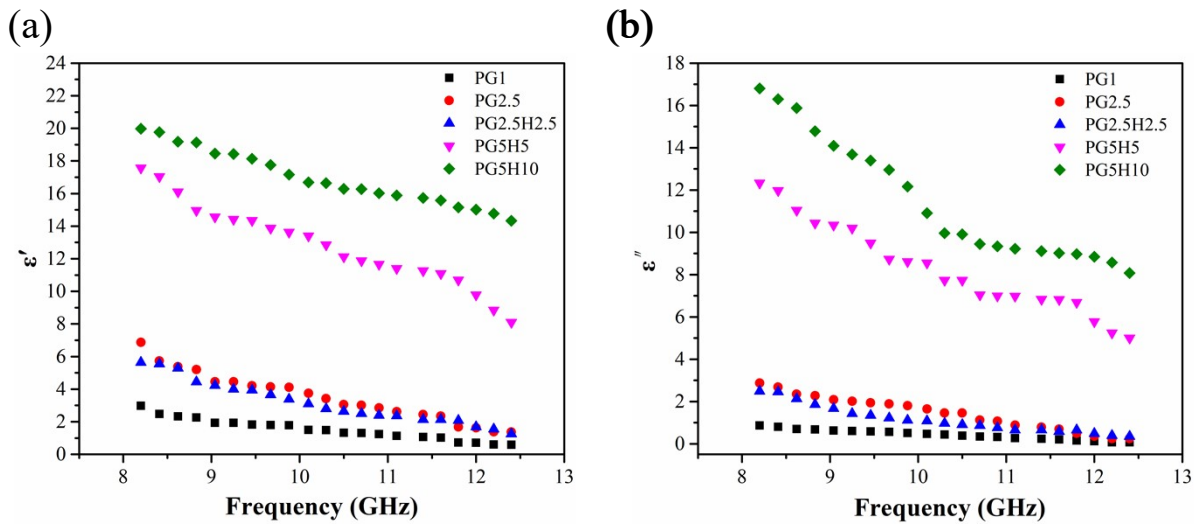

Figure S4. (a) ε′ and (b) ε″ of complex permittivity of the PG1, PG2.5, PG2.5H2.5, PG5H5, and PG5H10 nanocomposites.