

Supplementary Materials: Epitaxial Crystallization of Precisely Methyl-Substituted Polyethylene Induced by Carbon Nanotubes and Graphene

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Figure S1 shows the ¹H NMR spectra for the ADMET polymer PE21M and its precursors. The monomer is featured with the two signals of terminal alkenes appearing at 5.0 and 5.8 ppm (Figure S1C). The corresponding polymer (Figure S1B) exhibits signals of internal alkenes at 5.4 ppm. Meanwhile, signals of the terminal alkene are invisible in the ¹H NMR spectrum, indicative of the high molecular weight of the unsaturated polymer. After exhaustive hydrogenation of the unsaturated polymer, PE21M (Figure S1A) is characterized by complete disappearance of the signals of internal alkenes at 5.4 ppm. The ¹H NMR for PE21M shows a methine proton at 1.26 ppm, methylene protons at 1.08 ppm, and methyl protons at 0.85 ppm, a perfectly sequenced polymer, corresponding to polyethylene with methyl on every 21st backbone carbon.

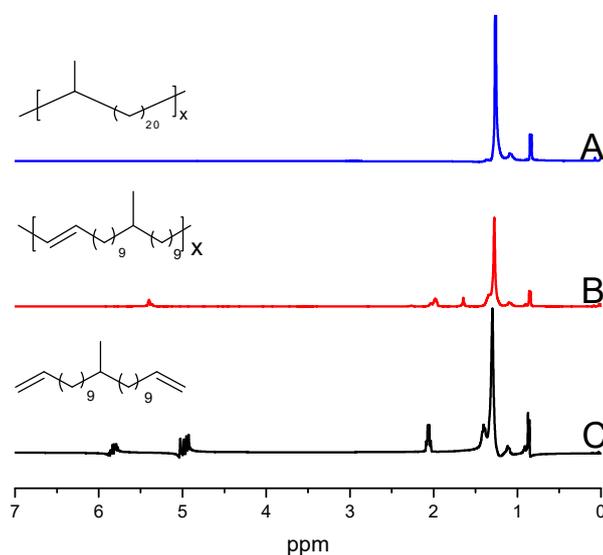


Figure S1. ¹H NMR for (A) PE21M, (B) unsaturated polymer, and (C) monomer.

As shown in Figure S2, two peaks centered at 19.4° and 22.2° were observed from PE21M/SWCNT nanocomposites prepared at 50 °C and 15 MPa SC CO₂ pressure, which can be indexed as (100) and (010) crystalline planes of triclinic form. The diffracted intensity of (100) and (010) crystalline planes is much stronger than that of PE21M/SWCNT

nanocomposite prepared by traditional solution crystallization, which suggests that supercritical CO₂ can effectively promote the growth of lamellae on SWCNT. The peak of 18.2° observed from PE21M/SWCNT nanocomposite is attributed to the SWCNT treated by SC CO₂ pressure. The reason why the peak of 18.2° appears in the diffractogram of SWCNT after treatment by SC CO₂ pressure is not yet clear. The positions of the diffraction peaks of PE21M/SWCNT nanocomposite prepared with the assistance of supercritical CO₂ are the same as that prepared by traditional solution crystallization. This indicates that supercritical CO₂ cannot change the crystal structure of PE21M/SWCNT composites.

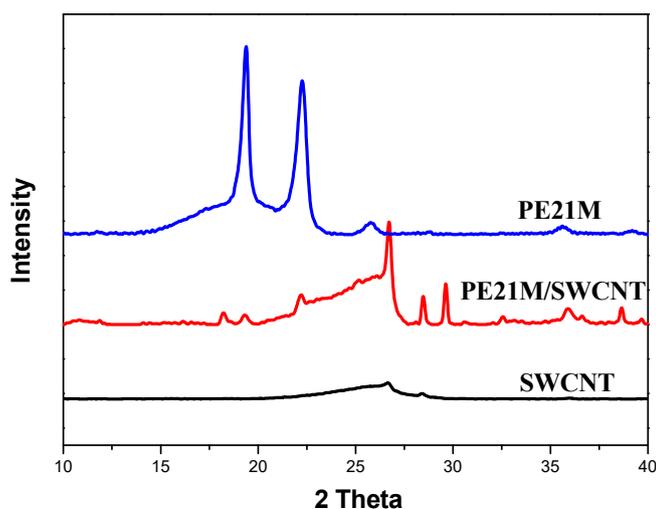


Figure S2. XRD diffractograms of PE21M and PE21M/SWCNT composite produced at 50 °C and 15 MPa SC CO₂ pressure.

As shown in Figure S3, two peaks centered at 19.4° and 22.2° were observed from PE21M/RGO nanocomposites prepared at 50 °C and 15 MPa SC CO₂ pressure, which can be indexed as (100) and (010) crystalline planes of triclinic form. The diffracted intensity of (100) and (010) crystalline planes is also much stronger than that of PE21M/RGO nanocomposite prepared by traditional solution crystallization. The same as with PE21M/SWCNT nanocomposite, the peak at 18.2° also appears in the diffractograms of RGO and PE21Br/RGO nanocomposite after treatment by SC CO₂ pressure. The positions of the diffraction peaks of PE21M/RGO nanocomposite prepared with the assistance of supercritical CO₂ are the same as that prepared by traditional solution crystallization. This indicates that supercritical CO₂ cannot change the crystal structure of PE21M/RGO composites.

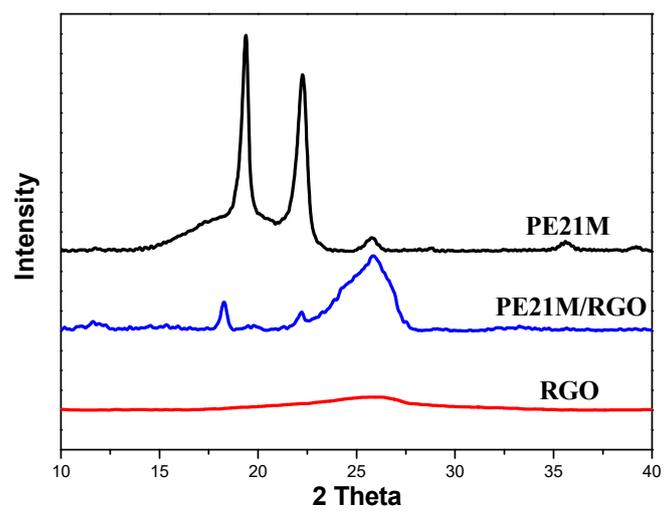


Figure S3. XRD diffractograms of PE21M and PE21M/RGO composite produced at 50 °C and 15 MPa SC CO₂ pressure.