Supporting info for:

Graphene Nanosheets induced Intrachain Conformational Ordering in a Semi-crystalline Polymer

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Experimental Section

Materials. iPP (Grade T30S) was provided by Dushanzi Petroleum Chemical Co., China, with a melt flow rate of 3 g/ 10 min (230 °C, 21.6 N), $M_w = 39.9 \times 10^4$ g mol⁻¹, and $M_w/M_n = 4.6$. Expandable graphites are used for the preparation of GNSs. The concentrated sulphuric acid (98 % H2SO4), hydrochloric acid (37 % HCl), potassium permanganate (KMnO4), hydrogen peroxide (30 % H2O2) were purchased from Chengdu Kelong Chemical Reagent Factory (China) as received.

The detailed preparation process of graphene nanosheets is described below.

Preparation of Graphene Nanosheets. Graphene nanosheets were synthesized from expanded graphites by the modified "Hummers" method in this study. The expanded graphite powders (20 g) were added into the concentrated H₂SO₄ (460 mL). Followed

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by slowly adding KMnO₄ (60 g) with stirring, so that the temperature of the mixture was not allowed to reach 20 °C. Then the suspension was stirred at 35 °C for 2 h. After that deionized water (920 mL) was poured in tardily and carefully. The reaction was terminated by the addition of a large amount of deionized water (2.8 L) and 30 %H₂O₂ solution (50 mL), causing violent effervescence and the temperature increased to almost 100 °C, as well as the color of the suspension changed to bright yellow. The suspension was washed by 1:10 HCl solution (5 L) in order to remove metal ions in filter papers and funnels. The paste collected from the filter papers was dried at 60 °C, until it became caking. Then the caking was dispersed into deionized water with stirring to reach static state and washed again by much deionized water for $5 \sim 7$ times in filter papers and funnels, until the PH of which on the filter papers reached nearly 7. Subsequently, the brown dispersion, namely graphene nanosheents was obtained by dispersed the paste collected on the filter papers into deionized water using ultrasonication and centrifugating at 4000 r.p.m. for 30 min to remove any unexfoliated graphite. At last the graphene nanosheets were obtained by dehydration of hydrosol at 60 °C in air.

Preparation of iPP/GNS Nanocomposites. Solution coagulation was utilized to prepare iPP/GNSs nanocomposites. In view of feasibly dispersion and highly efficient fabrication, N,N-dimethylformamide (DMF) was used as a solvent for GNSs. Taking iPP/0.1 wt % GNS nanocomposites as an example, the detailed experimental procedures showed below: 10 g of iPP was first dissolved in 100 mL of xylene at 130

°C by constant stirring in an oil bath. Then, 10 mg of GNSs were dispersed in 100 mL of DMF and sonicated for 15 min to obtain a uniform dispersion. By dropping the predispersed DMF/GNSs suspension into the xylene/iPP hybrid, the coagulations precipitated incessantly. After the above operation a mess of C_2H_5OH was poured into the mixture until no coagulations precipitated. Then the coagulations formed by iPP/GNSs nanocomposites were transferred to evaporating dishes overnight at room temperature and then dried in vacuum oven for 1 day at 80 °C to evaporate the any residual solvent. The films of iPP/GNS nanocomposites were obtained by molding the coagulations at 200 °C for 5 min.

Morphology of GONSs. The morphology of GNSs were examined by Nanoscope Multimode & Explore atomic force microscopy (Vecco Instruments, USA). Samples for AFM observation were prepared by depositing dispersions of GNSs in deionized water on a fresh Si substrate and allowing them to dry in the air. Typical images were obtained with line scan rates of 2 Hz and line scanning of 521 under ambient conditions

Figure S1 shows the AFM image of GNSs. It can be seen that the GNSs synthesized were only $1 \sim 2$ nm in thickness and their average length calculated was 0.87µm with a large distribution. The GNSs consist of only $1 \sim 3$ layers of graphites stacking in a separate platelet according to the thickness of one layer in the range of 0.8 ~ 1.2 nm.



Figure S1. AFM image of GNSs.

Dispersion of graphene nanosheets. The dispersion of graphene nanosheets (GNSs) was examined by a FEI Tecnai F20 with an acceleration voltage of 200 kV. Sample films of PPG05 were prepared with a thickness of 80 nm using a Leica EMUC6/FC6 microtome. Transmission electron micrograph of PPG05 is presented in Figure S1. It can be observed that the GNSs are well exfoliated and some smaller sheets are also seen due to the ultrasonic disruption.



Figure S2. Transmission electronic micrograph of GNSs in iPP/GNS nanocomposites.

Differential scanning calorimetry (DSC) measurements. DSC measurements of iPP/GNS nanocomposites were carried out on a TA Instruments' Q200 under a nitrogen flow, which was calibrated by indium as the standard. For non-isothermal crystallization, samples (~ 5 mg) were first annealed at 180 °C for 5 minutes to erase any thermal history and then cooled to 80 °C at a cooling rate of 10 °C/min. For isothermal crystallization, the samples were annealed at 180 °C for 5 min to eliminate thermal history, cooled to desired crystallization temperature (*T*c) at a cooling rate of 20 °C/min, and maintained at *T*c until the crystallization was completed. The exothermal traces were recorded for the later data analysis.



Figure S3. DSC exothermal curves of PPG0, PPG05, and PPG10 at a cooling rate of 10 °C/min.



Figure S4. Heat flow curves (a) and relative crystallinity (b) of PPG10, PPG05, and PPG0 during isothermally crystallizing at 138 °C.