**Supplementary Information:**

**Experimental**

Poly (propylene carbonate) (PPC) pellets was supplied by Changchun Institute of Applied Chemistry, Chinese Academy of Science. Its weight-average molecular weight (Mw) was $2.48 \times 10^5$ g/mol and Mw/Mn was 3.2. GO sheets were prepared by the oxidation of natural graphite using the Hummers method.\(^{30}\) PPC/GO nanocomposites with 0-1 wt% GO were then prepared as follows (Scheme 1): concentrated and well-dispersed GO in water was firstly prepared with the aid of ultrasonication. 10g PPC was dissolved in 100mL THF under vigorous stirring to yield a clear solution. Then a certain amount of GO/H\(_2\)O solution was added into the PPC/THF solution. The concentration of the GO inside the PPC matrix was quantitatively controlled by syringe titration. After that, this homogeneous PPC/GO mixture was gradually poured into distilled water with stirring. After coagulation in large amount of water, PPC/GO nanocomposites powder was collected, repeatedly washed and dried until its weight unchanged. The composites were then compression molded for testing at a temperature of 130°C.

Tensile testing of neat PPC and PPC/GO composites was determined using an Instron testing machine at 23°C with a crosshead speed of 100mm/min. The tensile specimens were cut with a length of mm, a width of 4 mm and a thickness of 1mm. In order to ensure accuracy and repeatability, at least 8 specimens were tested.

The tensile-fractured surfaces of the specimens were observed using an Inspect field-emission SEM (FEI Company, USA) with 5 kV accelerating voltage.
Dynamic mechanical analysis was performed using a dynamic mechanical analyzer (DMA Q800, TA instrument). A heating rate of 3°C/min and a frequency of 1Hz were employed under multifrequency-strain mode. At least two tests were carried out for each case.
Fig. S1 Schematic representation of novel solution mixing process illustrating the dispersion of GO in the PPC matrix.
**Fig. S2** Tensile fracture morphologies of the neat PPC and PPC/GO nanocomposites.

(a) neat PPC. (b) PPC/0.1wt%GO. (c) PPC/0.7wt%GO. (d) PPC/1wt%GO.

With the increasing content of GO, the increased fibrillar-like network of PPC is observed after tensile deformation. GO sheets in the PPC matrix could act as effective physical cross-linking points to hinder PPC chain deformation. This behavior is similar to the cross-linking of natural rubber by sulfur, which leads to a sharply improvement of the tensile strength and modulus.