

Green dielectric materials composed of natural graphite minerals and biodegradable polymer

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S1. SEM micrographs of natural graphite minerals after ultrasonic processing. From Figure S1, natural graphite minerals contained flakes with a diameter of about 5-20 μm .

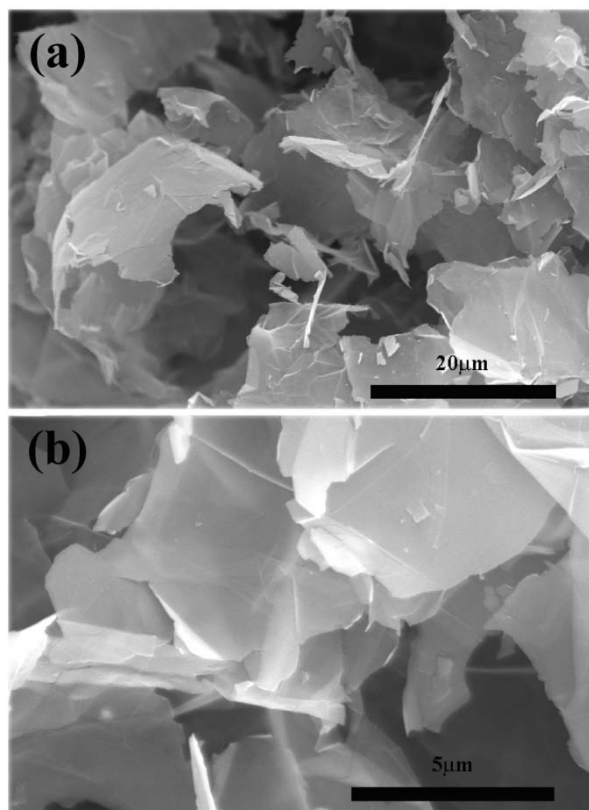


Figure S1. SEM micrographs of natural graphite minerals after ultrasonic processing.

S2. XRD of natural graphite minerals after ultrasonic processing. From Figure S2, a 002 diffraction peak at $2\theta = 26.48$ indicated that natural graphite minerals after ultrasonic processing had a multilayered structure composed of about 20-60 carbon layers. This was confirmed by Figure S1(b). (Ref. Fuan He, Sienting Lau, *Adv. Mater.*, **2009**, 21, 710).

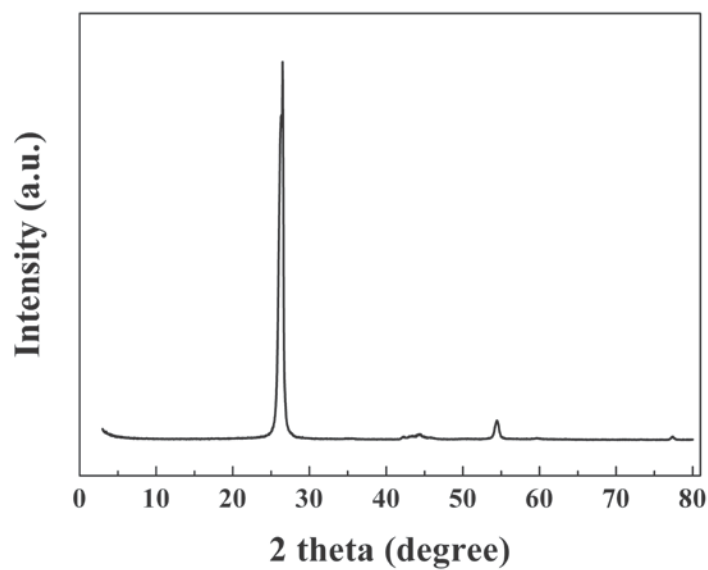


Figure S2. XRD of natural graphite minerals after ultrasonic processing.

S3. Schematic illustration of NNGM/PBS composite films formation: (i) NNGM was firstly dispersed in the solution by ultrasonic treatment and rigorously stirred with PBS matrix. (ii) The solution was casted on glass and composite films were fabricated by compressing molding several times.

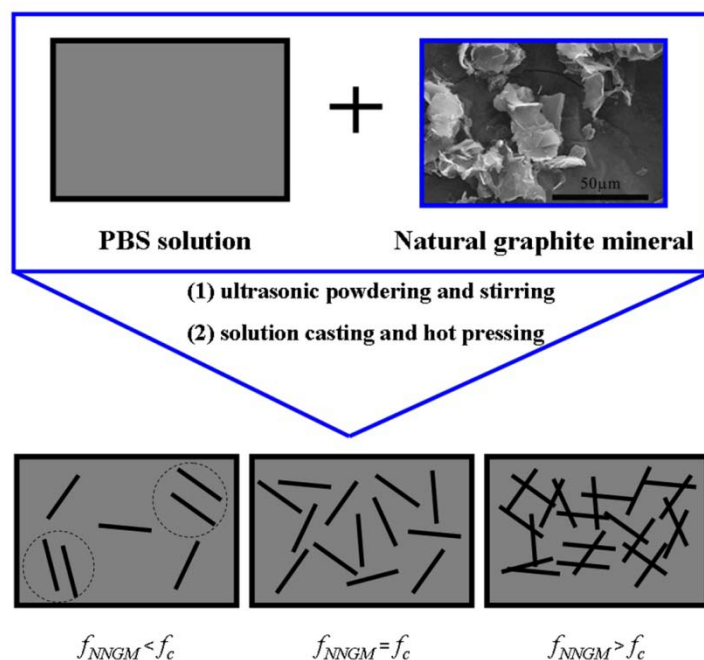


Figure S3. Schematic illustration of NNGM/PBS composite films formation.

S4. Raman results of NNGM and NNGM/PBS composite films with 2.5 % NNGM:

For pristine NNGM, the peak at 1339.6 cm^{-1} assigned to disorder-induced D band from the multiple photo scattering of defects or amorphous carbon. The peak located at 1579.1 cm^{-1} assigns to G band assigns to the stretching of conjugated double bonds corresponding to sp^2 hybridization. For NNGM/PBS composite films with 2.5 %, D band shifts was attributed to compressive forces associated with polymer chains on NNGM (Ref. J. K. Yuan, S. H. Yao, Z. M. Dang, *J Phys. Chem. C* **2011**, 115, 5515).

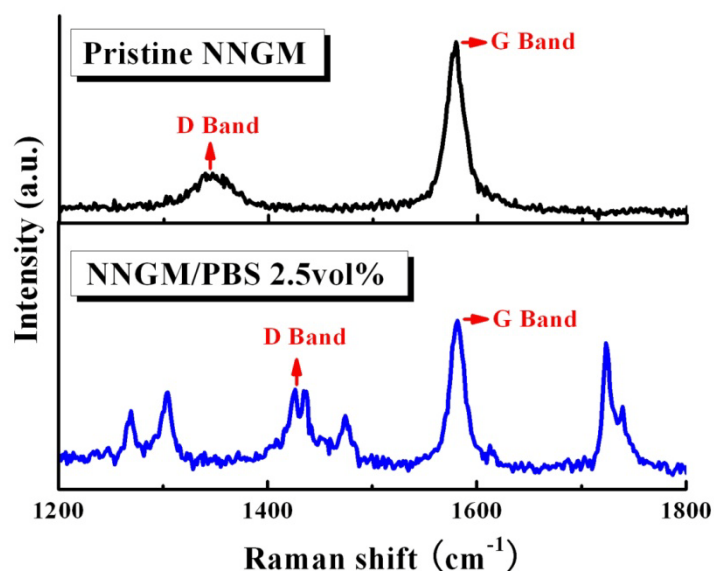
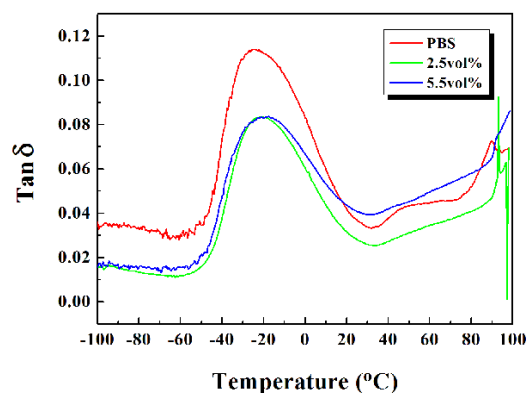


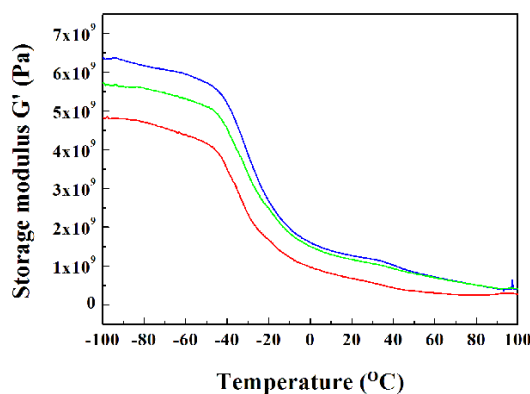
Figure S4. Raman results of NNGM and NNGM/PBS composite films with 2.5 %.

S5. DMA results of PBS and NNGM/PBS composite films with 2.5 % and 5.5 %:

From Figure S5(a), $\tan\delta$ peak of pure PBS at around $-23\text{ }^{\circ}\text{C}$ corresponded to the glass-transition temperature. For NNGM/PBS composite films, $\tan\delta$ peak moved toward a high temperature due to the hindering action between natural graphite minerals and polymer chain indicated interaction between natural graphite minerals. Similar results have been reported for polyamide/clay nanocomposites by Tjong (Ref. S. C. Tjong, S. P. Bao, *J Polym Sci Part B*, **2004**, 42, 2878.). From Figure S5(b), G' were $4.2 \times 10^3\text{ MPa}$ and $6.4 \times 10^3\text{ MPa}$ for pure PBS and NNGM/PBS composite films with 5.5 %, respectively.



(a)



(b)

Figure S5. DMA results of PBS and NNGM/PBS composite films with 2.5 % and 5.5 %.