Supporting Information

3D Printing of Polyethylene Terephthalate Glycol–Sepiolite Composites with Nanoscale Orientation

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Supplementary materials

Sepiolite (SEP) was purchased from Sigma-Aldrich (USA). PETG or poly(ethylene glycol-co-1,4-cyclohexanedimethanol terephthalate) was obtained as PN200 from SK Chemicals.

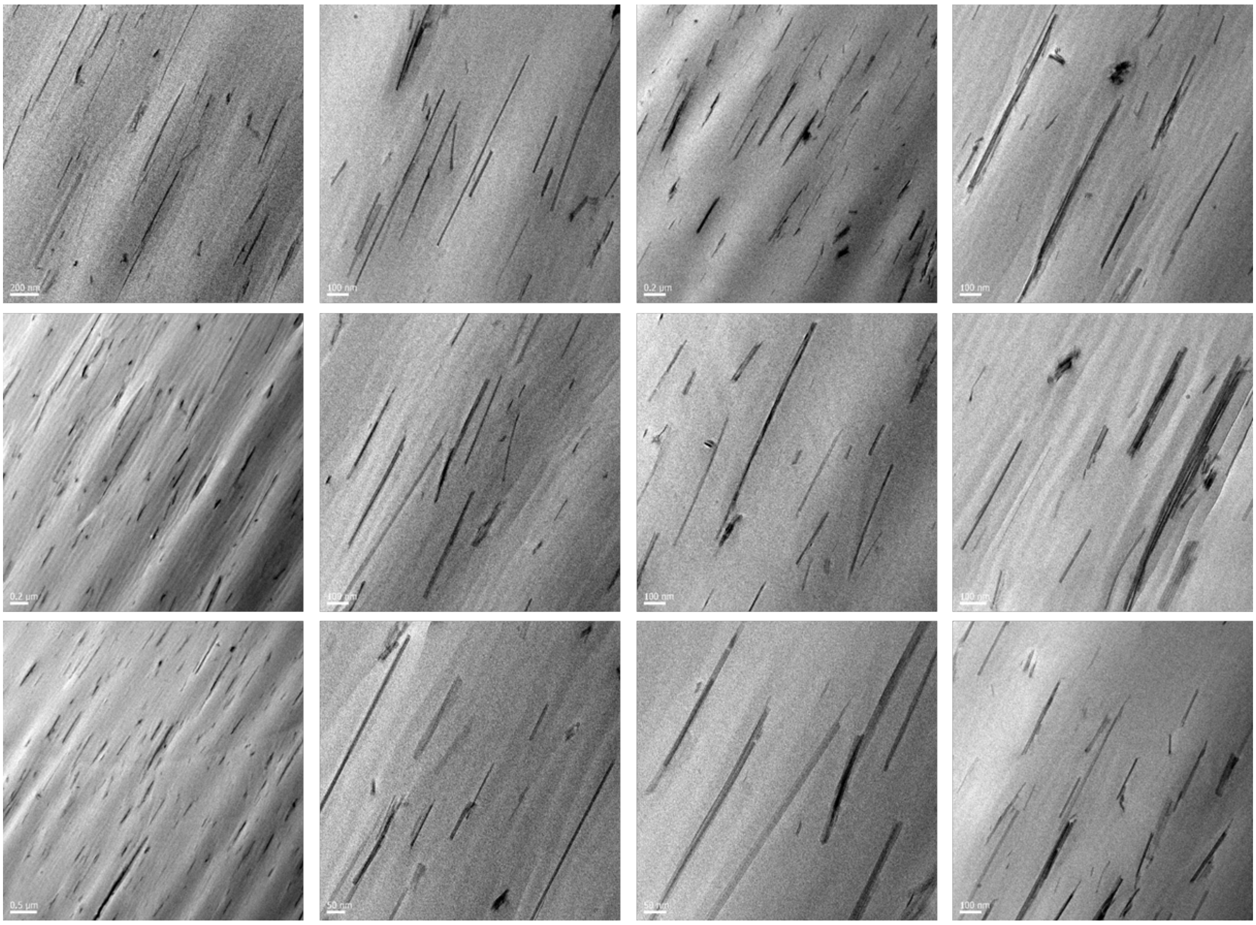

Figure S1. TEM images show the oriented sepiolite (SEP) structures in the 3D-printed PETG matrix of the SEP-3 composite.

The sample was observed by TEM as shown in Figure S1, different parts of the same sample were imaged. It was confirmed that the SEPs were all oriented in the same direction.

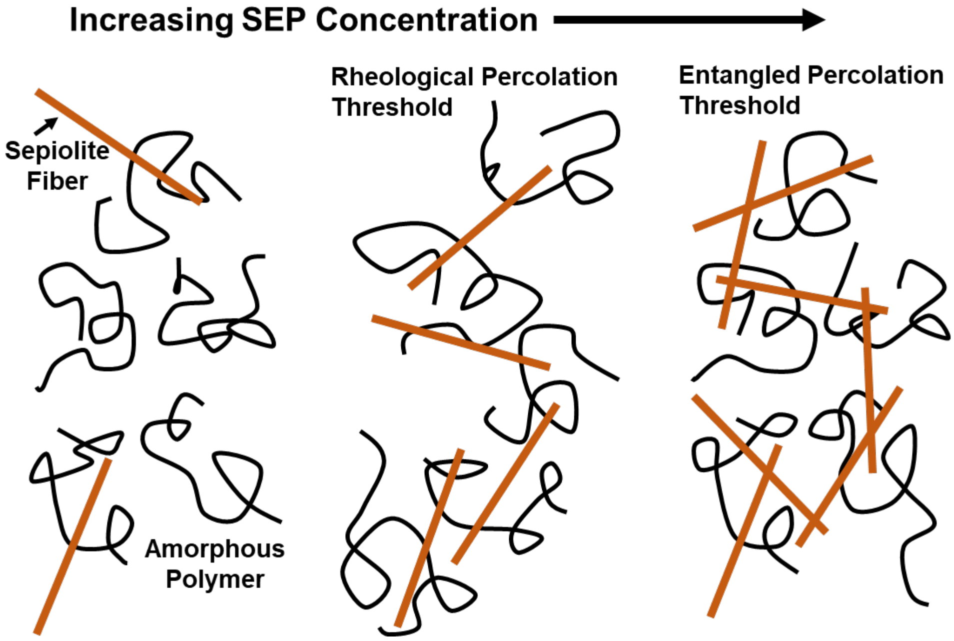

Figure S2. Schematic of PETG–SEP nanocomposites in which SEP has isotropic orientation. (left) For low SEP concentrations, the rheological properties of the composite are comparable to those of the host polymer. (middle) The onset of solid-like viscoelastic behavior occurs when the size of the polymer chain is somewhat large relative to the separation between SEP bundles. (right) Entanglement of SEP with PETG occurs. Reprinted with permission from ref 1. Copyright 2004 American Chemical Society.

The schematic in Figure S2 accounts for the phenomenon of PETG-SEP composites showing shear thinning in the molten state (as observed in Figure 1). In this study, we confirmed the phenomenon of shear thinning in SEP of more than 5phr.

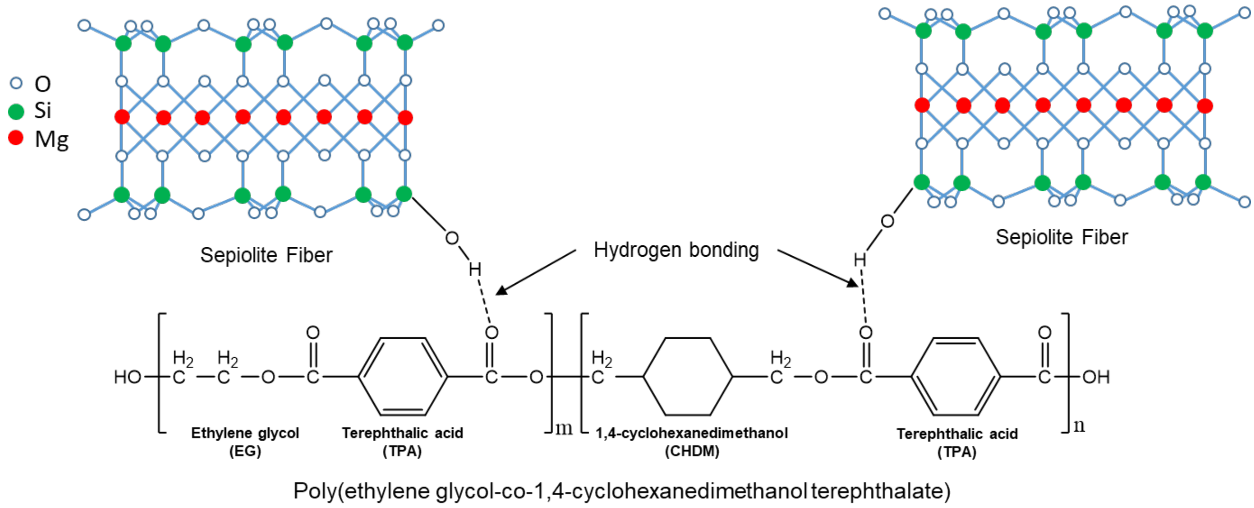

Figure S3. Schematic of the chemical bonding forces between PETG and SEP in PETG–SEP composite

Figure S3 is a schematic of the expected hydrogen bond formation between PETG and SEP. It explains the compatibility of PETG and SEP, demonstrating the reason that chemical treatment is unnecessary.

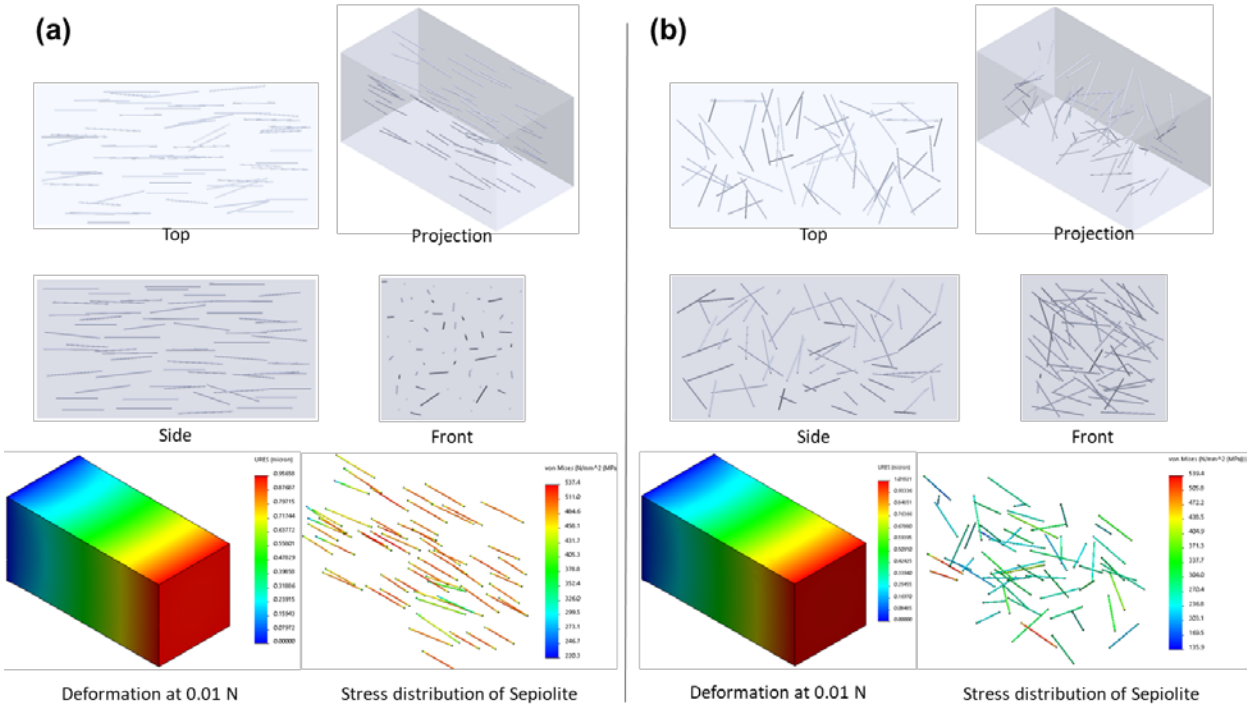

Figure S4. According to the fabrication methods of (a) 3D printing and (b) injection molding, different SEP structures are formed inside the composite, and the stress applied to SEP according to the structure is visualized through simulation.

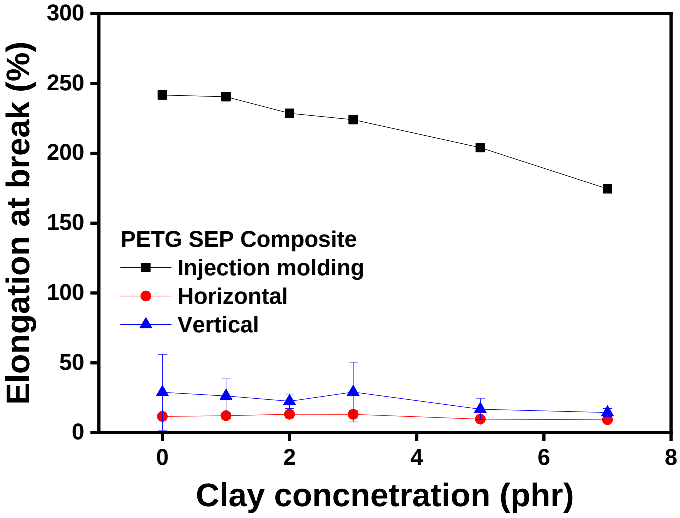

Figure S5. Mechanical property measurements of the elongation at break of PETG nanocomposites with SEP at varying filler contents.

This graph is associated with Figure 2b and c. The 3D-printed samples perform poorly under stretch deformation because the printing nozzles create defects on the surface during 3D printing.

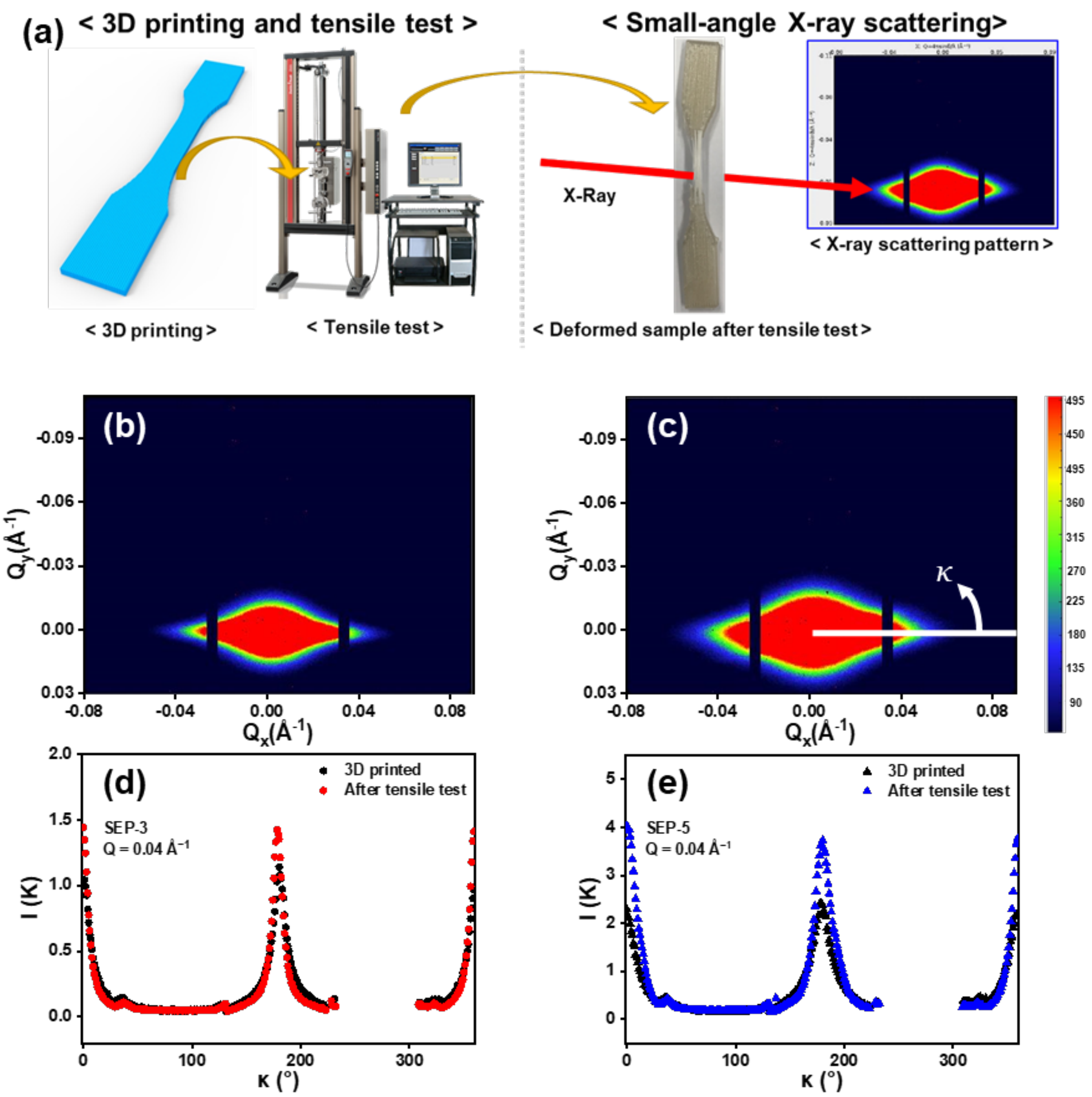

Figure S6. Sample preparation method and experimental results for SAXS measurement. (a) Schematic of 3D printing, tensile test, and SAXS measurement. SAXS data from 3D printed tensile sample after test of (b) SEP-3 composite, (c) SEP-5 composite. Azimuthal scattering intensity before and after tensile test of (d) SEP-3 composite, (e) SEP-5 composite at q = 0.04 $Å^{-1}$.

Reference

(1) Du, F.; Scogna, R. C.; Zhou, W.; Brand, S.; Fischer, J. E.; Winey, K. I. Nanotube Networks in Polymer Nanocomposites: Rheology and Electrical Conductivity. *Macromolecules* **2004,** *37* (24), 9048-9055.