Supporting Information

Self-limiting Selective Phase Separation of Graphene Oxide and Polymer Composites Solution

Feifan Chen[1], Lidan Wang[1], Kaiwen Li[1], Rui Guo[1], Yicong Qing[1], Chenwei Shen[1], Yingjun Liu[1,2], Zhen Xu[1,2], and Chao Gao,[1,2*]*

[1]MOE Key Laboratory of Macromolecular Synthesis and Functionalization, Department of Polymer Science and Engineering, Key Laboratory of Adsorption and Separation Materials & Technologies of Zhejiang Province, Zhejiang University, 38 Zheda Road, Hangzhou 310027, China.

[2]Shanxi-Zheda Institute of Advanced Materials and Chemical Engineering, Taiyuan 030032, China.

*Corresponding author. Email: zhenxu@zju.edu.cn (Z. X.); chaogao@zju.edu.cn (C. G.).

Table of contents

Figure S1. The state of different polymers dissolved in different solvents.

Figure S2. GO stacking process.

Figure S3. TEM image of SPS-GO/PS sample.

Figure S4. Morphology comparison of SPS-GO/PS sample before and after immersion in EA.

Figure S5. The process of PS migration and fusion.

Figure S6. Statistic distribution of diameter of PS with different concentration.

Figure S7. The CLSM image of SPS-GO/PS.

Figure S8. Statistic of maximum cluster area with different α_{PS} .

Figure S9. The method of the calculation of continuous phase ratio.

Figure S10. Water contact angle measurement of pure GO, PS, PEO, PVA and CNC.

Figure S11. The OM image of GO/PEO.

Figure S12. The DLS Intensity-Radius curve of pure GO solution and GO/PEO solution.

Figure S13. The DLS Count Rate-Time curve of pure GO solution and GO/PEO solution.

Figure S14. Scheme of the continuous SPS-spinning process.

Figure S15. Fracture section of homogeneous GO/PEO fibers.

Figure S16. TGA of SPS fibers and homogeneous fibers.

Figure S17. FTIR of pure GO, SPS fibers and homogeneous fibers.

Figure S18. GO/PEO solution precipitated in EA and EA/ethanol.

Figure S19. TGA of pure GO, SPS-GO/PEO and Homogeneous-GO/PEO.

Figure S20. Tensile curves of homogenous GO/PEO fibers and the SPS fibers.

Figure S21. SEM images of SPS-GO/PEO fibers with different type of knots.

Figure S22. Stress-strain curves of SPS fibers with different numbers of knots.

Table S1. The contact angle difference between different polymers and pure GO.

Table S2. The raw data for mechanical testing of homogenous fibers and SPS fibers.

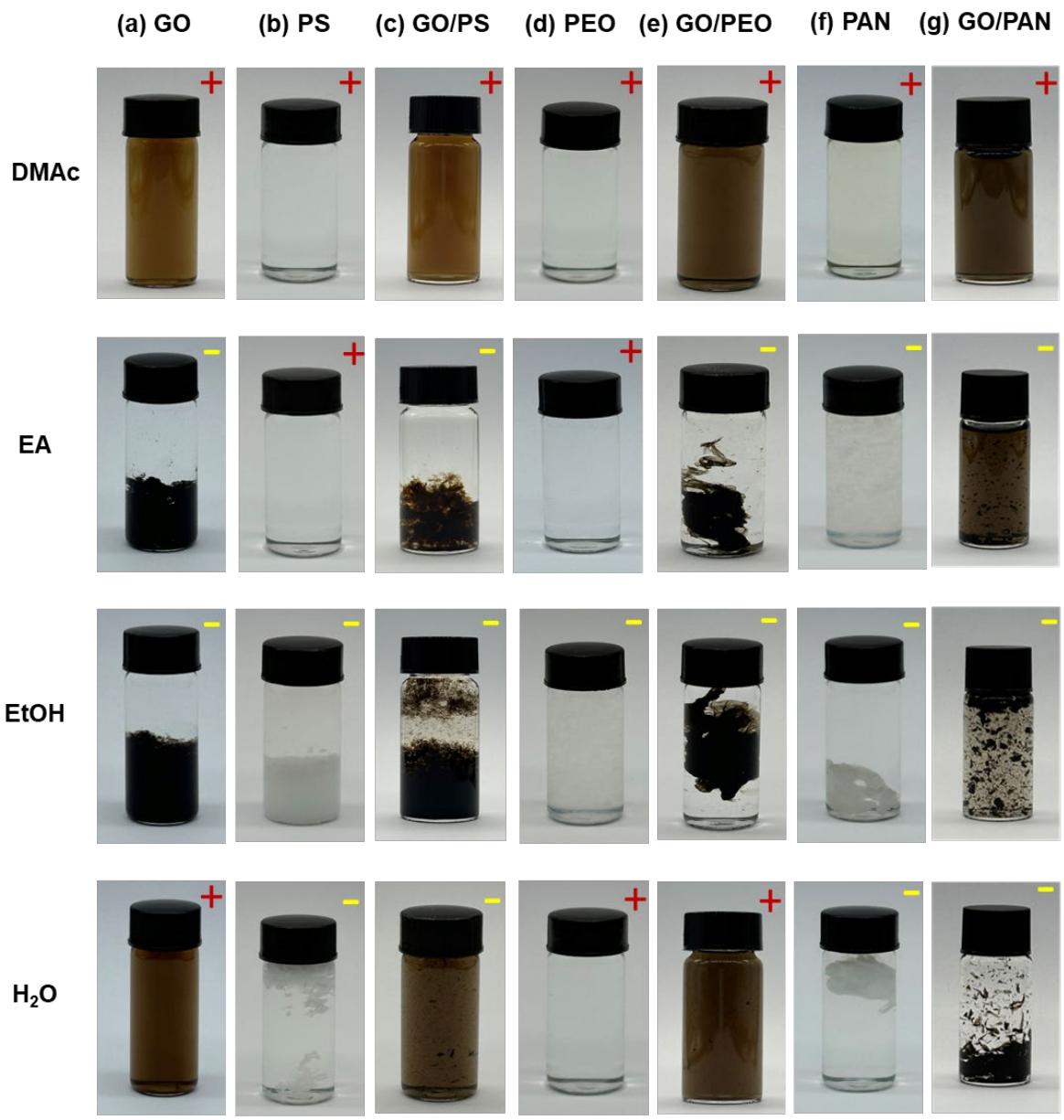

Figure S1. The state of different polymers dissolved in different solvents. (a) GO dispersion. (b) PS solution with Mw=280 kDa. (c) GO/PS (M_{GO}:M_{PS}=1:1) mixed solution. (d) PEO solution with Mw=7900 kDa. (e) GO/PEO (M_{GO}:M_{PEO}=1:1) mixed solution. (f) PAN solution with Mw=280 kDa. (g) GO/PAN (M_{GO}:M_{PAN}=1:1) mixed solution. "—", poor solvent. "+", good solvent.

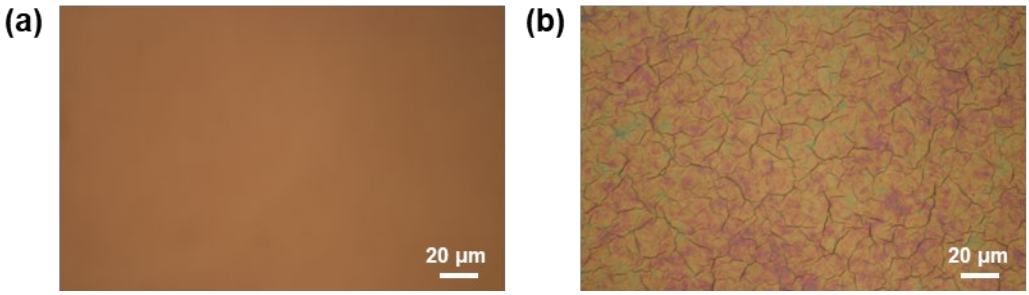

Figure S2. GO stacking process. (a) OM image of GO. (b) OM image of GO after immersion in EA.

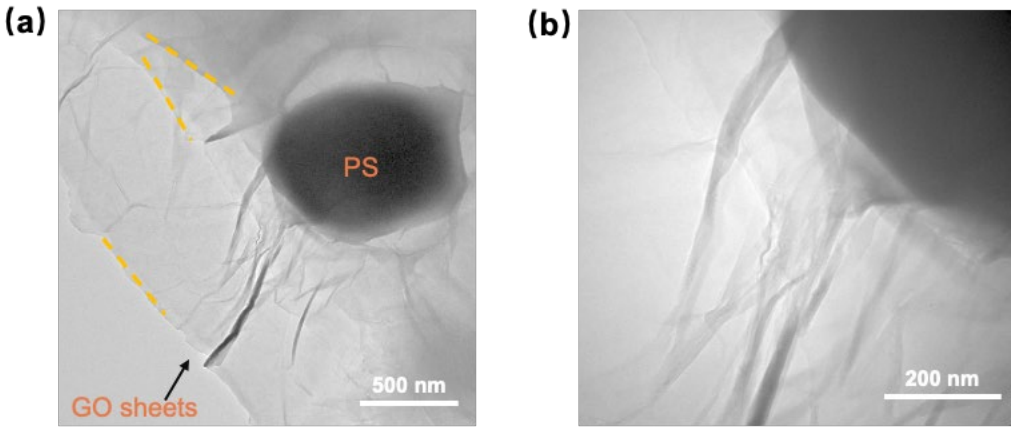

Figure S3. TEM image of SPS-GO/PS sample. The black sphere is PS which is covered with GO sheets.

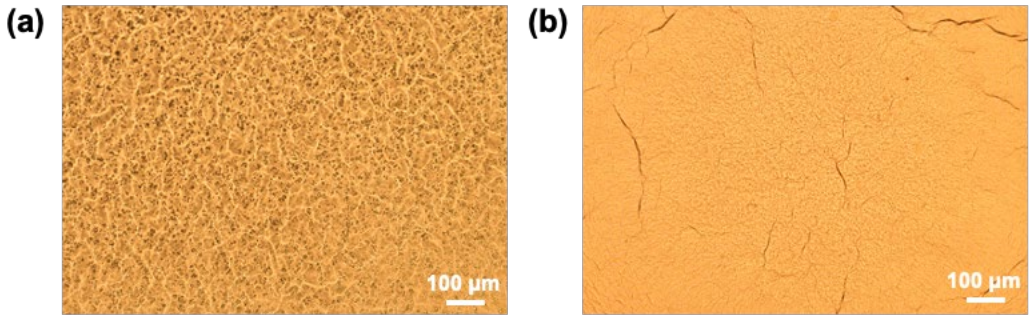

Figure S4. Morphology comparison of SPS-GO/PS sample before and after immersion in EA. (a) OM image of SPS-GO/PS sample before immersion in EA. (b) OM image of SPS-GO/PS sample after immersion in EA.

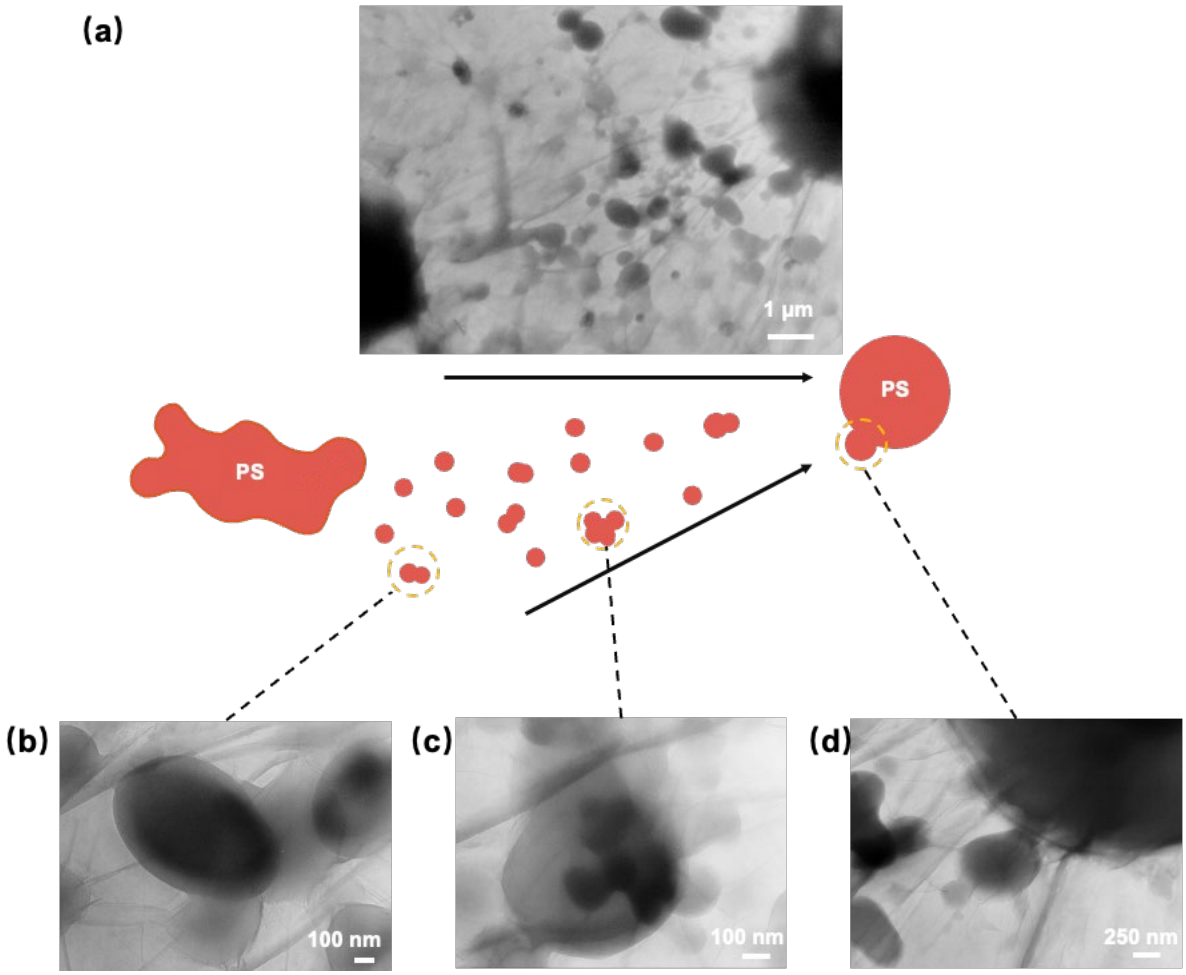

Figure S5. The process of PS migration and fusion. PS migrates and converges between layers to form a larger scale PS phase.

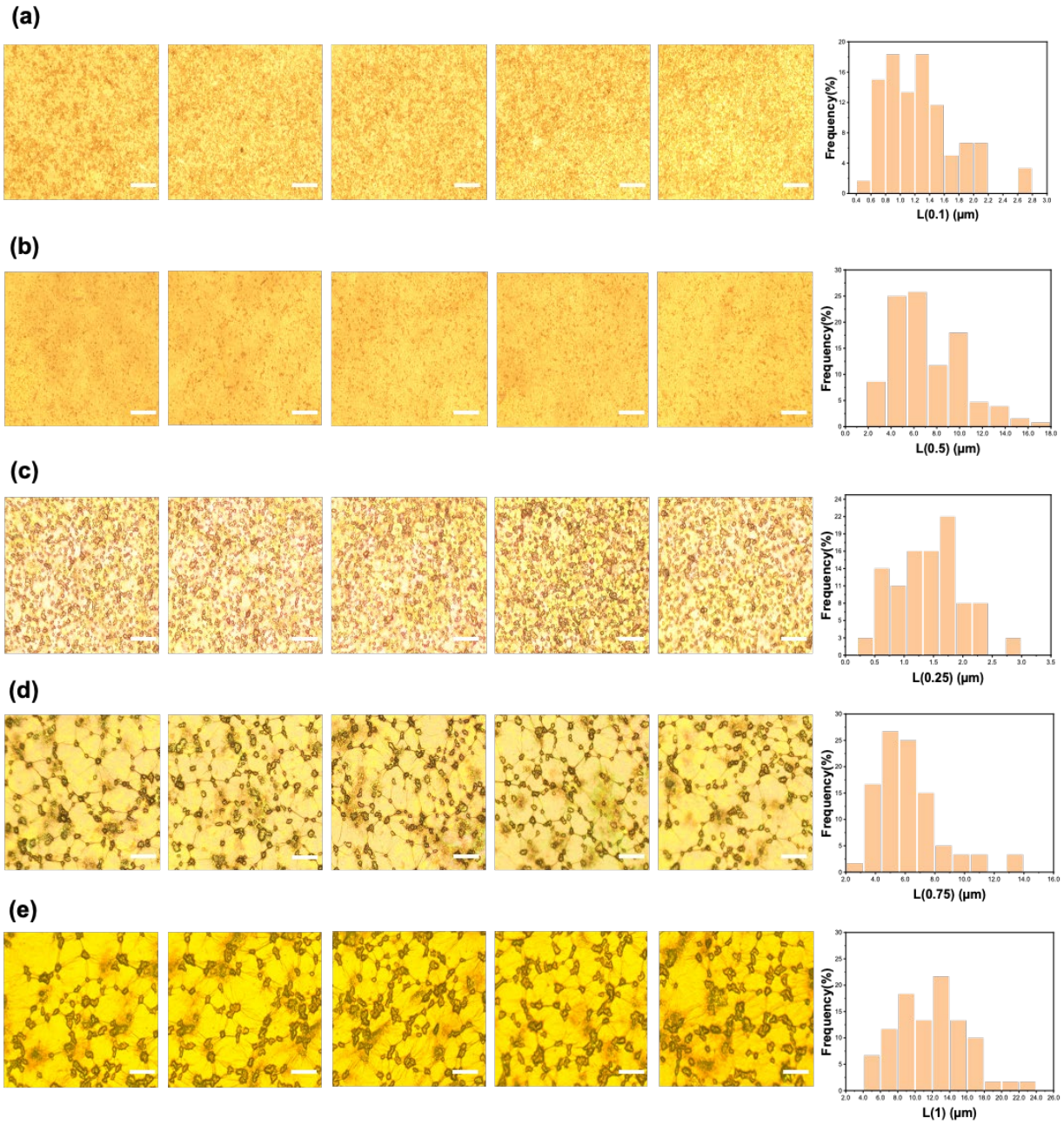

Figure S6. Statistic distribution of diameter of PS with different concentration. (a) 0.1%. (b) 0.25%. (c)0.5%. (d) 0.75%. (e) 1%. Scale bar is 20 μm.

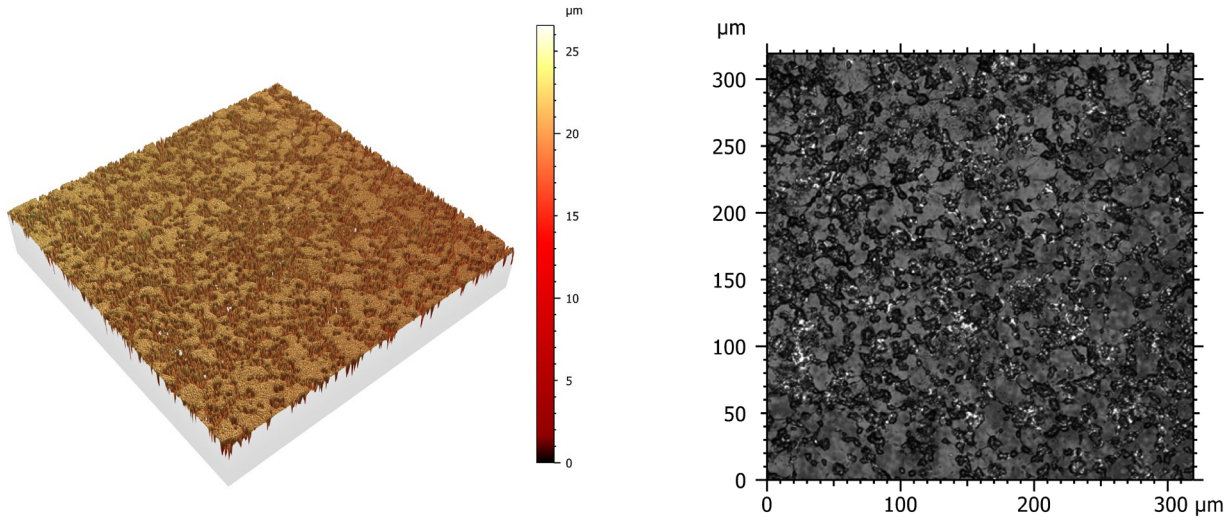

Figure S7. The CLSM image of SPS-GO/PS.

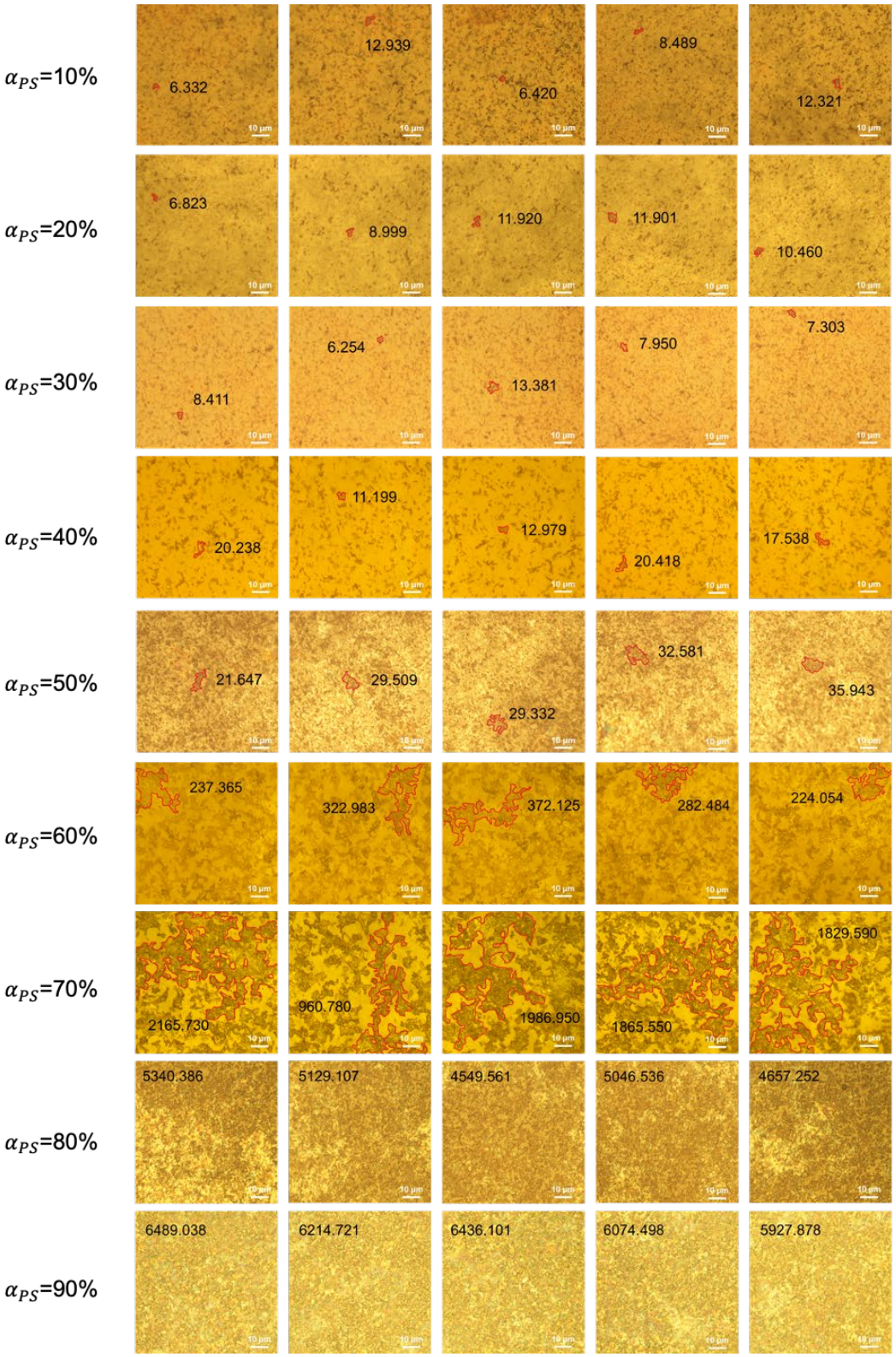

Figure S8. Statistic of maximum cluster area with different α_{PS} .

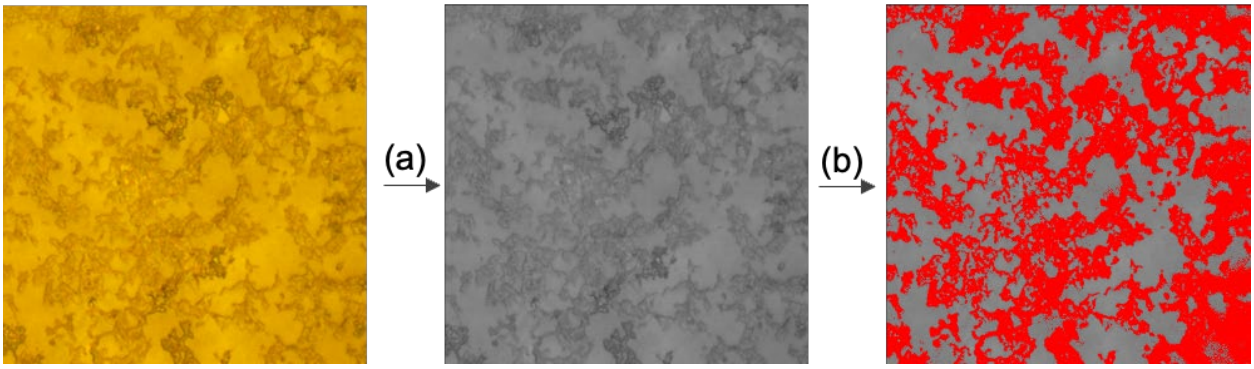

Figure S9. The method of the calculation of continuous phase ratio. (a) Change image into an 8-bit grayscale type. (b) Adjust threshold to select the phase separation area. The continuous phase ratio can be calculated: $\alpha = \frac{S_{contineous\ phase\ area}}{S_{area}}$.

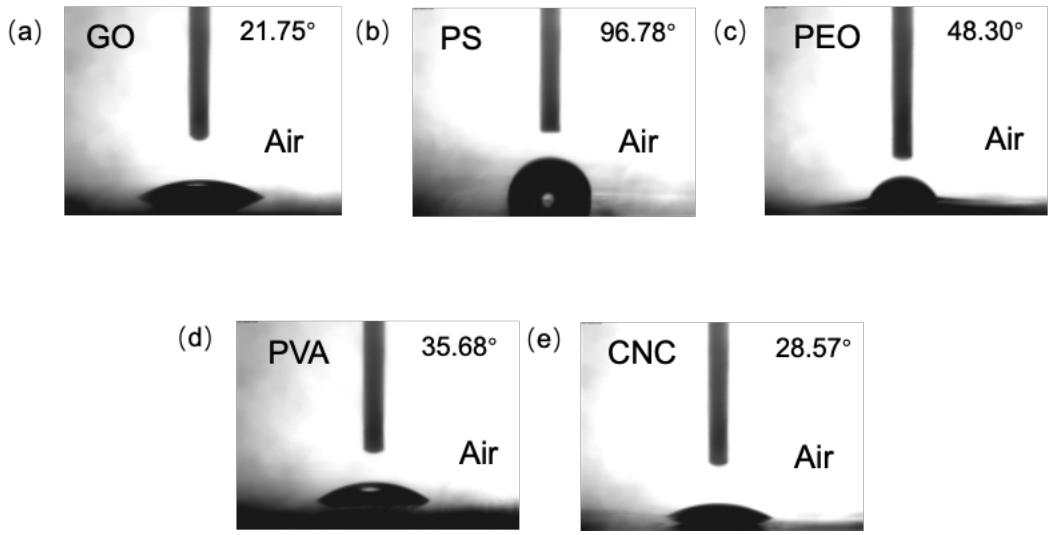

Figure S10. Water contact angle measurement of pure GO, PS, PEO, PVA and CNC.

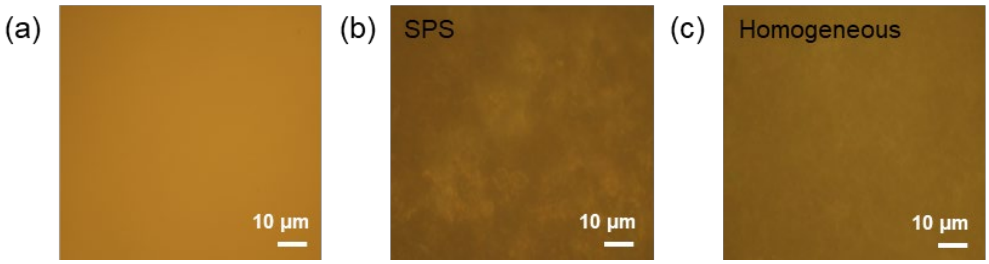

Figure S11. The OM image of (a) homogeneous solution, (b) after adding EA to generate SPS and (c) after adding EA/ ethanol to generate homogeneous aggregation for GO/PEO.

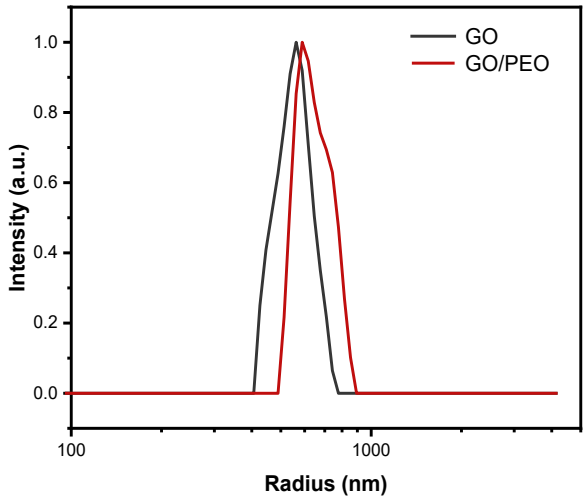

Figure S12. The DLS Intensity-Radius curve of pure GO solution and GO/PEO solution. The hydrodynamic radius of GO and GO/PEO is 562 nm and 588 nm.

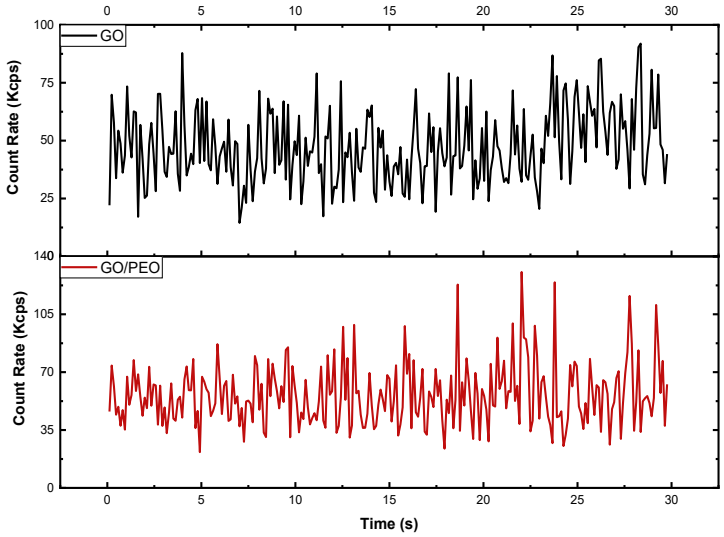

Figure S13. The DLS Count Rate-Time curve of pure GO solution and GO/PEO solution.

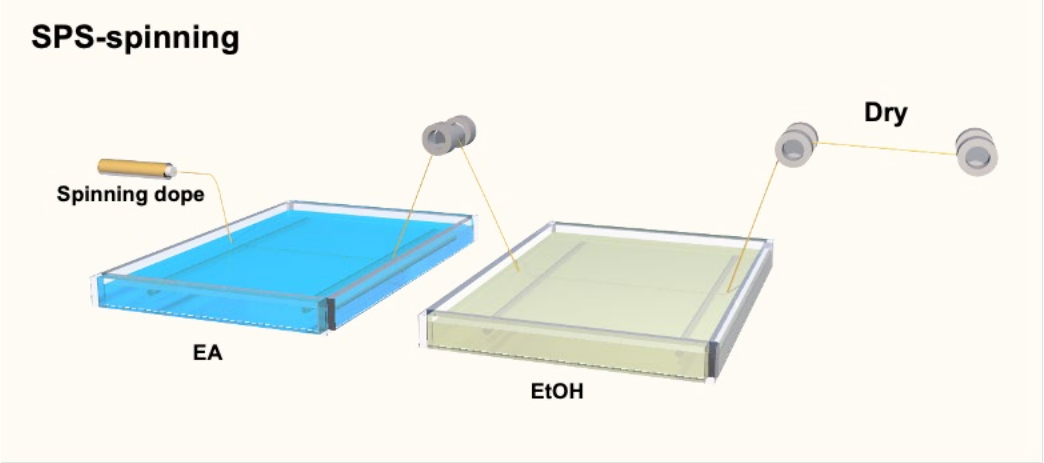

Figure S14. Scheme of the continuous SPS-spinning process.

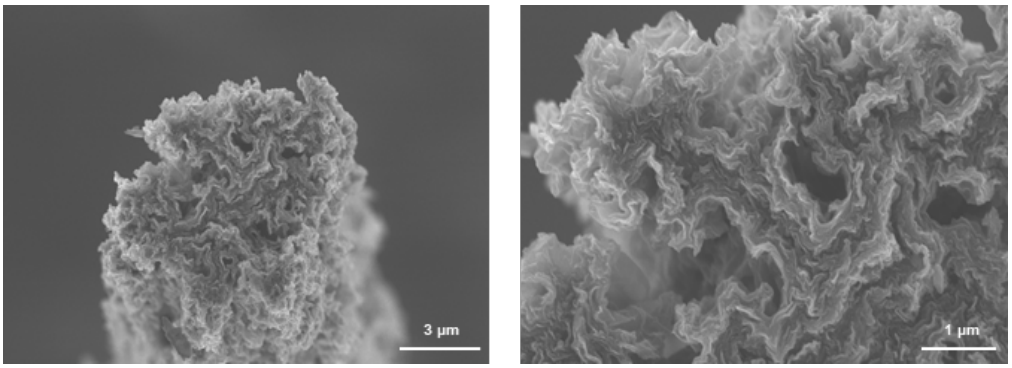

Figure S15. Fracture section of homogeneous GO/PEO fibers.

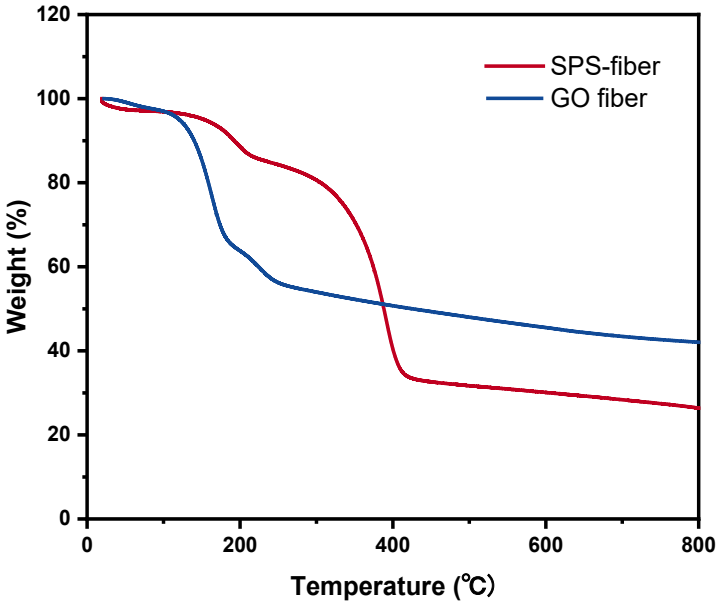

Figure S16. TGA of SPS fibers and GO fibers.

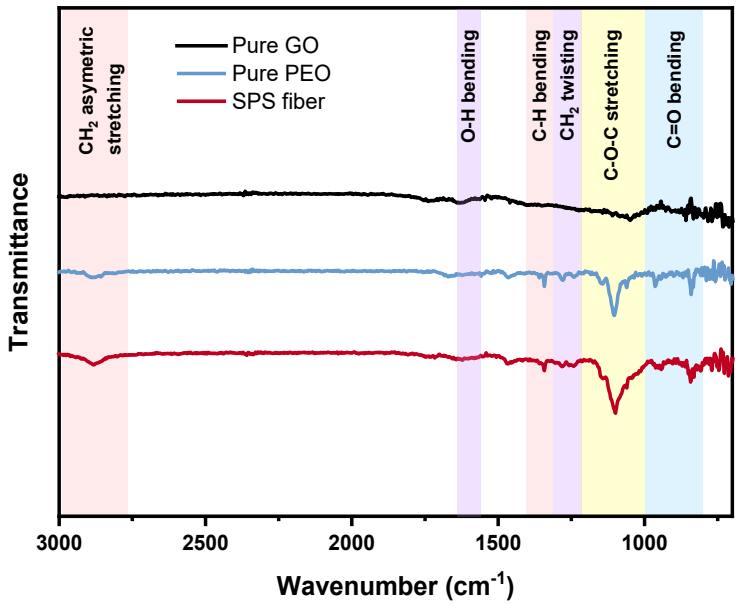

Figure S17. FTIR of pure GO, pure PEO and SPS fibers.

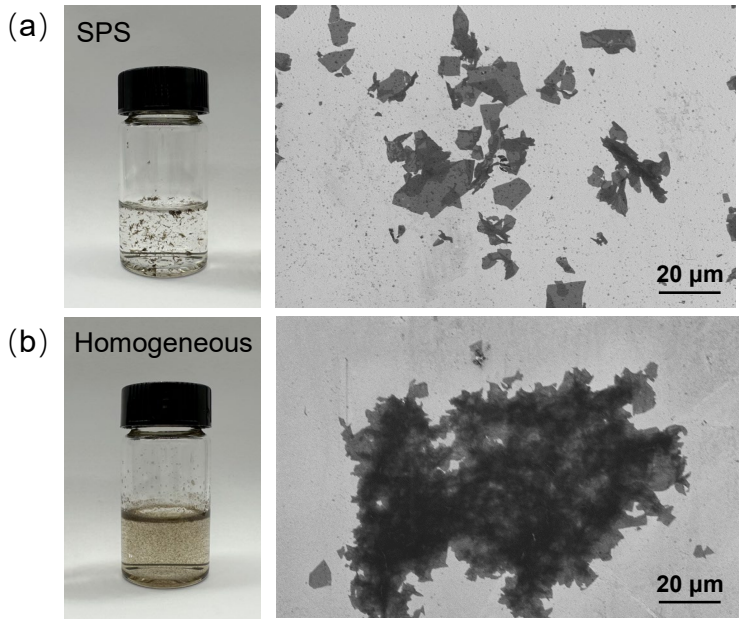

Figure S18. GO/PEO solution precipitated in EA and EA/ethanol. (a) Photo of GO/PEO solution precipitated in EA, followed by repeatedly washing by EA. The right panel is SEM image of the aggregated GO sheets. (b) Photo of GO/PEO solution precipitated in EA/ethanol, followed by repeatedly washing by EA/ethanol. The right panel is SEM image of the aggregated GO sheets with PEO.

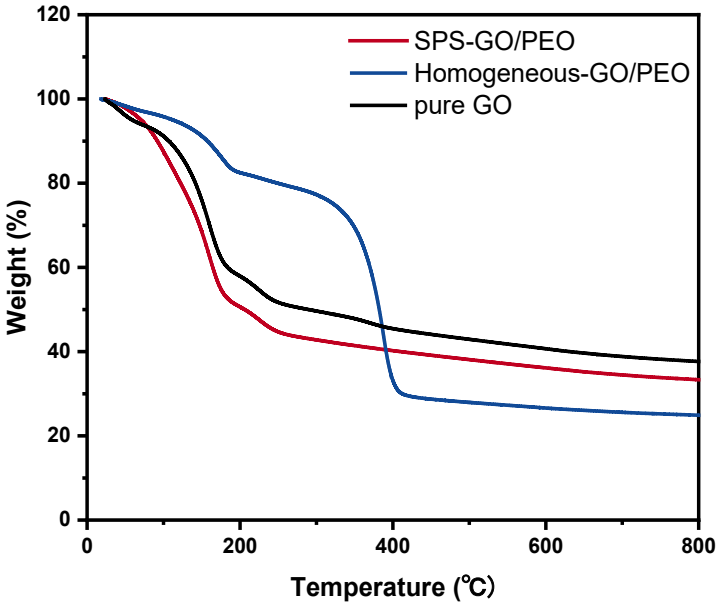

Figure S19. TGA of pure GO, SPS-GO/PEO and Homogeneous-GO/PEO.

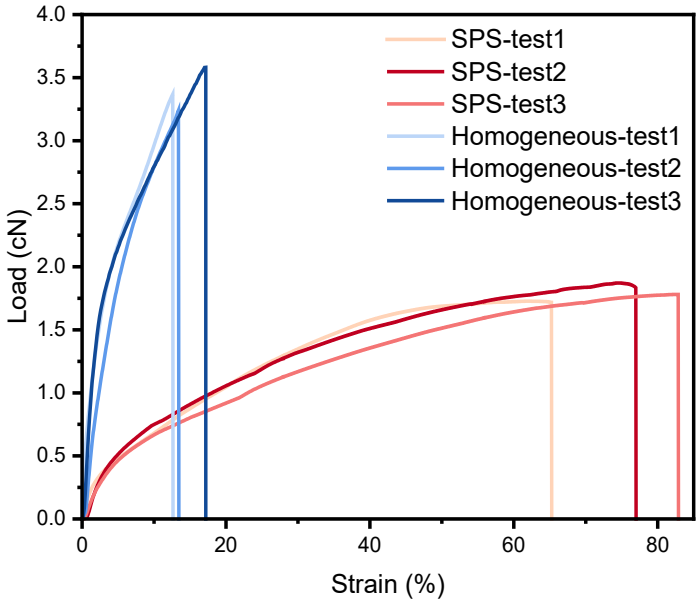

Figure S20. Tensile curves of homogenous GO/PEO fibers and the SPS fibers.

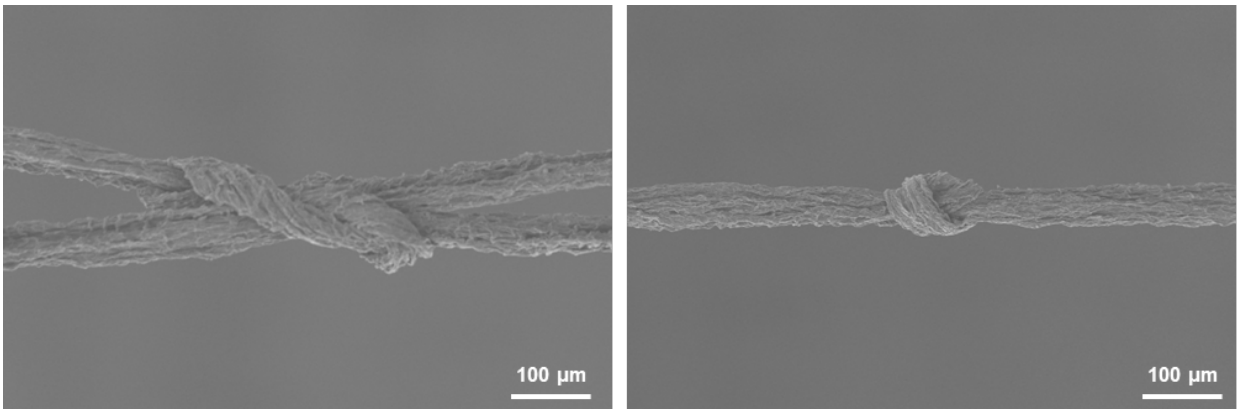

Figure S21. SEM images of SPS fibers with different type of knots.

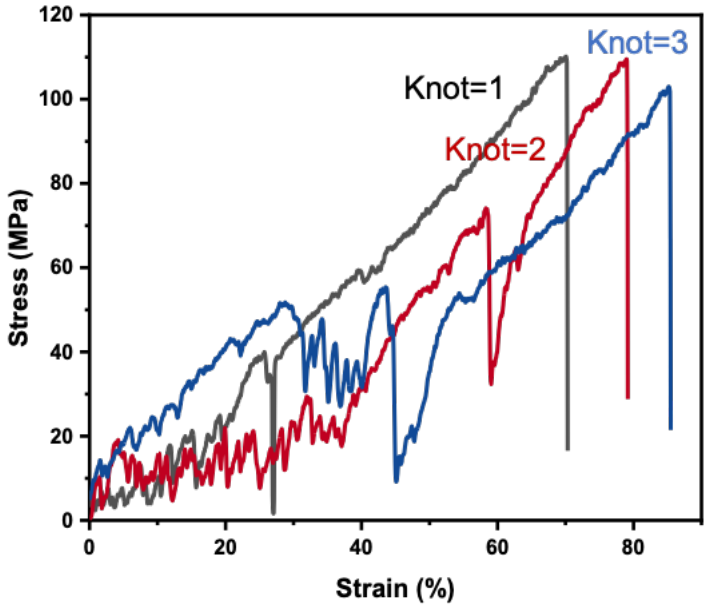

Figure S22. Stress-strain curves of SPS fibers with different numbers of knots.

Table S1. The contact angle difference between different polymers and pure GO.

	PS	**PEO**	**PVA**	**CNC**
Δθ (°)	75.03	26.55	13.92	6.82

Table S2. The raw data for mechanical testing of homogenous fibers and SPS fibers.

Sample	Strain (%)	Load (cN)
SPS-test1	65.28	1.72729
SPS-test2	76.96	1.87142
SPS-test3	82.88	1.78035
Homogenous-test1	12.64	3.36892
Homogenous-test2	13.36	3.23955
Homogenous-test3	17.16	3.58193