Supplementary Information (SI) for Green Chemistry.

Innovative Green Synthesis of Hydrophobic Covalent Networks Using Ethyl Cellulose/Thymol Eutectic Systems

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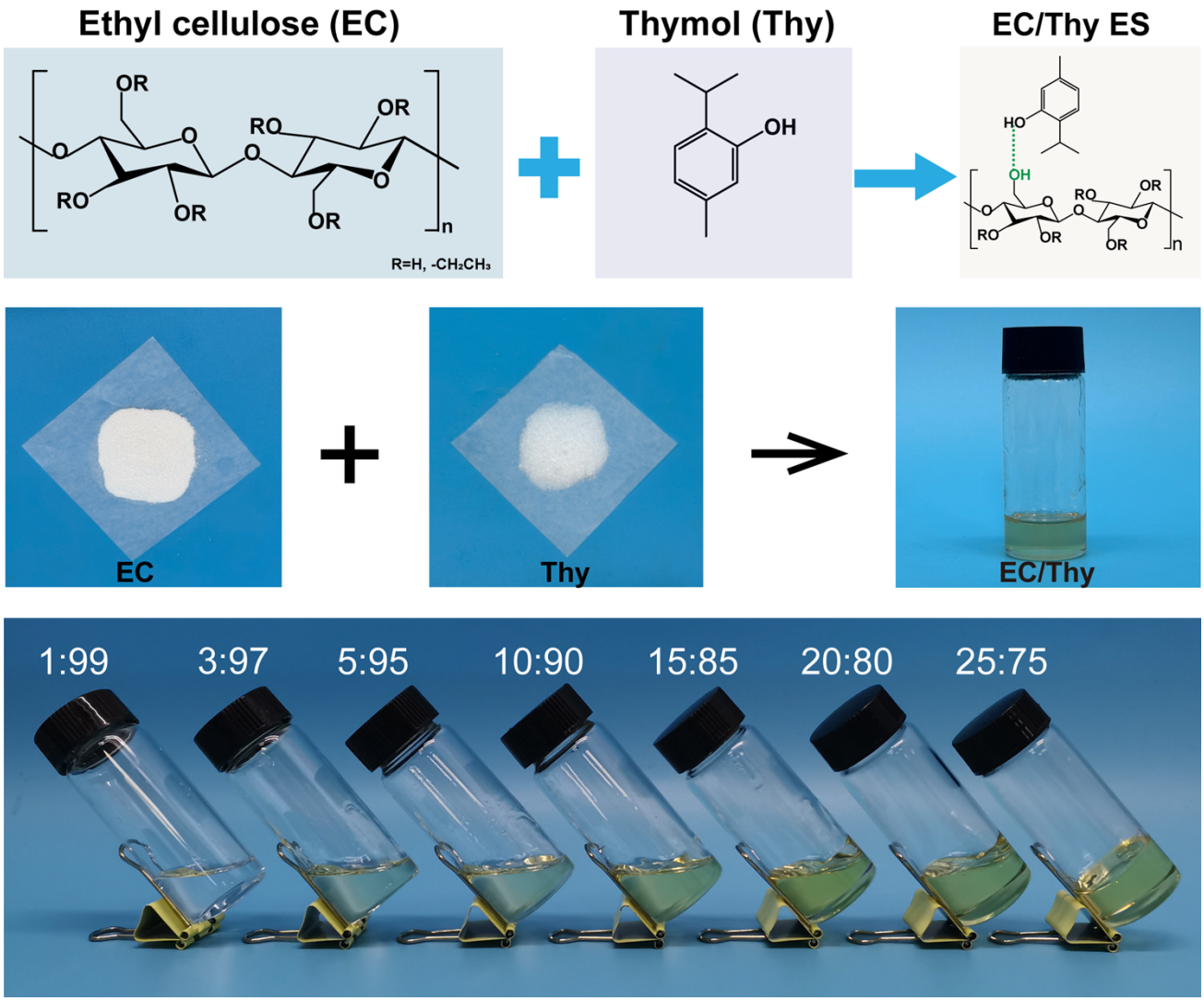

Figure S1. EC and Thy can form clear and transparent solutions within a wide mass ratio range. As the content of EC in the mixed solution increases, the viscosity continuously rises.

Table S1. Molecular weight information of EC from different manufacturers

Sample	**Mn**	**Mw**
EC-1 (Tianjin Zhonglian Chemical Reagent)	65421	68664
EC-2 (Aladdin Reagents)	75402	76030
EC-3 (Shanghai yuanye Bio-Technology)	269760	461142
EC-4 (Tianjin Huasheng Chemical Reagent)	65011	68660
EC-5 (Shanghai Macklin)	77700	78185

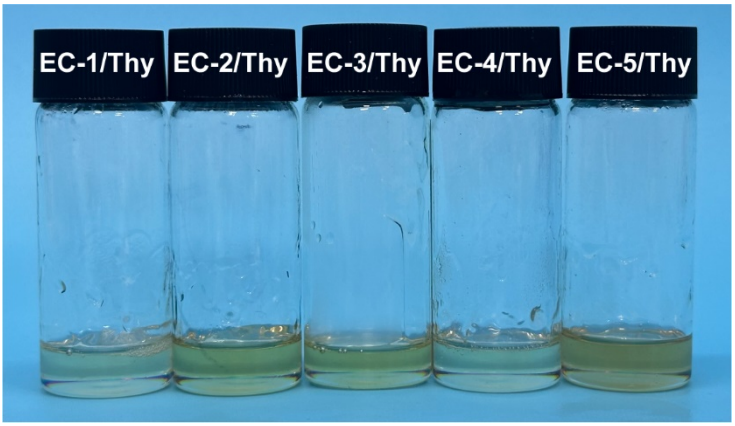

Figure S2. EC/Thy mixtures prepared with EC from different manufacturers.

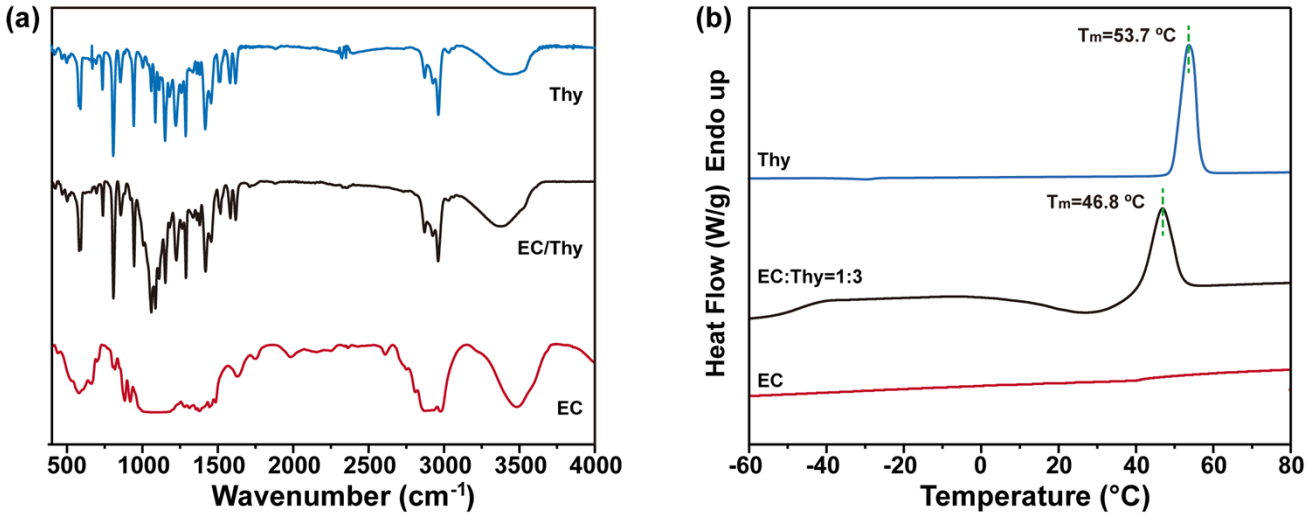

Figure S3. (a) FTIR spectra and (b) DSC curves of EC, Thy, and EC/Thy ES. From the FTIR spectrum, it can be seen that no chemical reactions occurred during the preparation process. The formation of the EC/Thy mixture mainly relies on intermolecular hydrogen bonding interactions. The DSC curve of the EC/Thy mixture reveals that the melting point of the mixture is significantly lower than the melting points of either EC or Thy alone. The depression of the melting point is mainly attributed to the hydrogen bonding interactions between the components.

Table S2. The physicochemical properties of the EC/Thy ES

Sample	Water content	pH	Viscosity (25°C)
EC:Thy=1:3	0.92 wt%	6.55	450 Pa.s

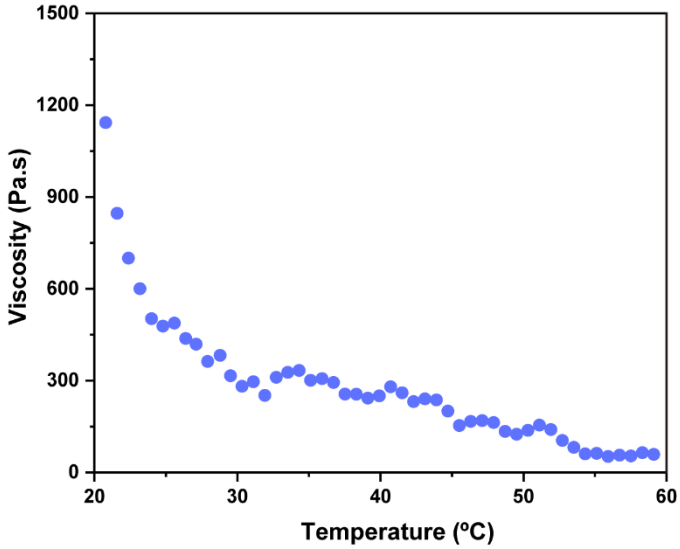

Figure S4. The viscosity of EC/Thy ES (1:3) varies with temperature.

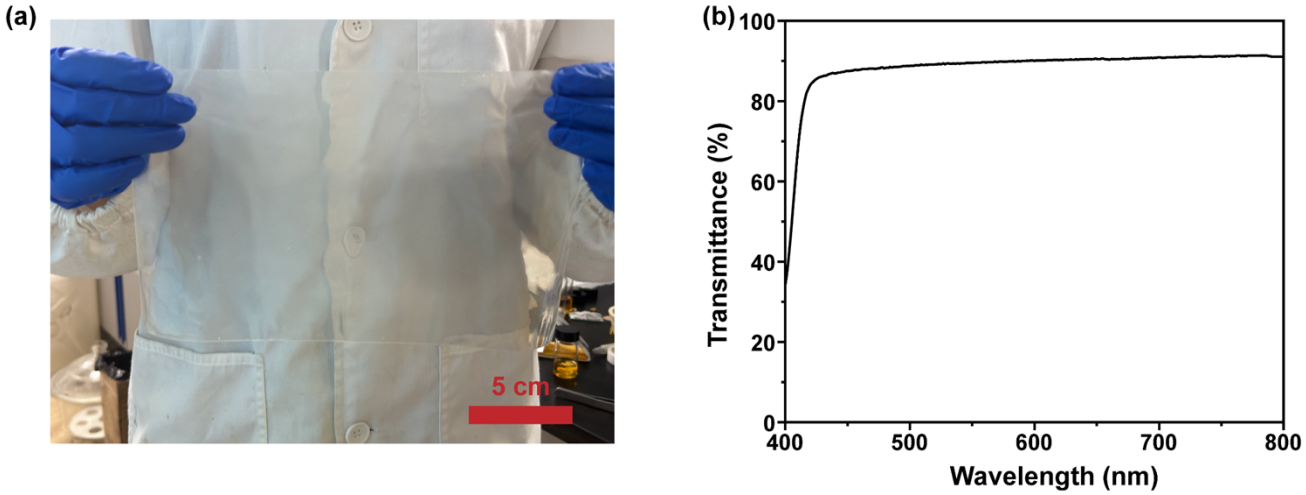

Figure S5. (a) The CPF with a large format exhibits high transparency. (b) The optical transmittance of CPF in the visible light region.

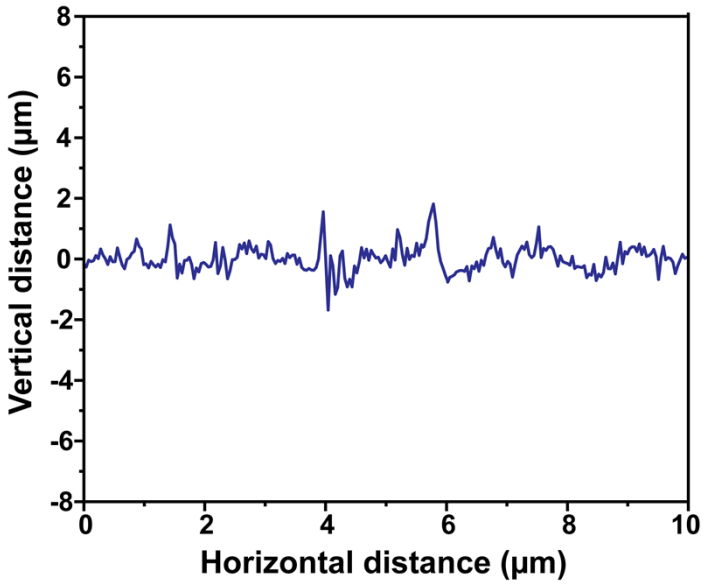

Figure S6. The surface roughness of CPF obtained by AFM. The result shows that the surface of CPF fluctuates very little and has a very high smoothness.

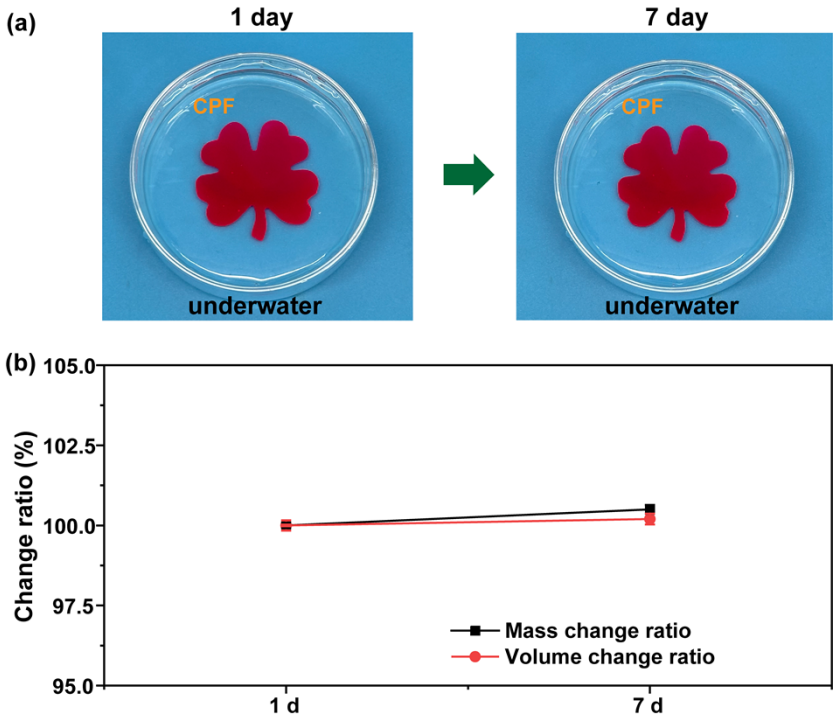

Figure S7. (a) Optical photographs of CPF after soaking underwater for different periods of time. (b) The mass and volume change ratios of the prepared CPF when soaked underwater for 1 day and 7 days

To measure volume changes, the differences in length (L), width (W) and thickness (T) of the samples before and after immersion were calculated.

$$Volume\ change\ ratio = \frac{V_{new} - V_{original}}{V_{original}} \times 100\%$$

$$V = l \times w \times t$$

To measure mass changes, the changes in mass of the samples before and after immersion were determined.

$$Mass\ change\ ratio = \frac{M_{new} - M_{original}}{M_{original}} \times 100\%$$

All samples were wiped with an absorbent cloth to remove any residual moisture from the surface prior to measurement.

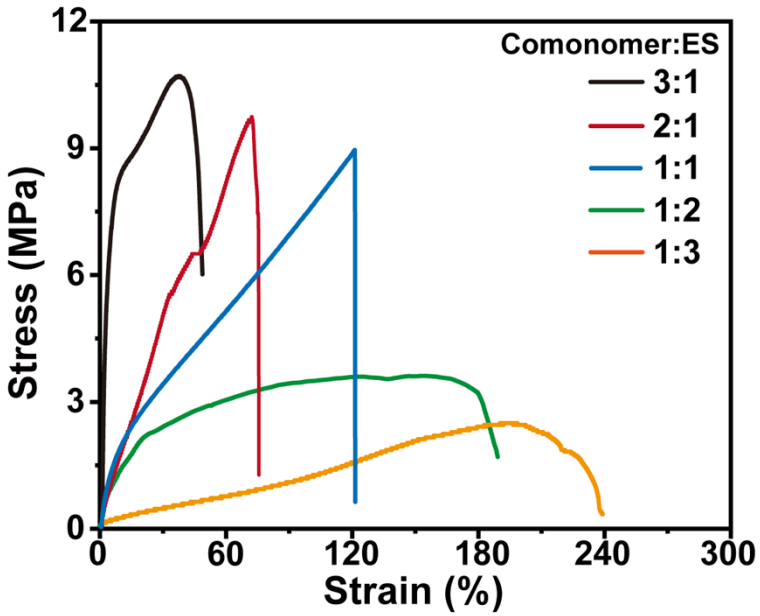

Figure S8. The mechanical properties of CPFs prepared with different ratios of comonomers to ES. For samples with higher comonomer content (comonomer: ES = 3:1 or 2:1), excessive chemical crosslinking leads to higher mechanical strength and lower tensile deformation of the synthesized CPFs. With increasing ES content (comonomer: ES = 1:2 or 1:3), insufficient chemical crosslinking significantly reduces the mechanical properties of CPFs.

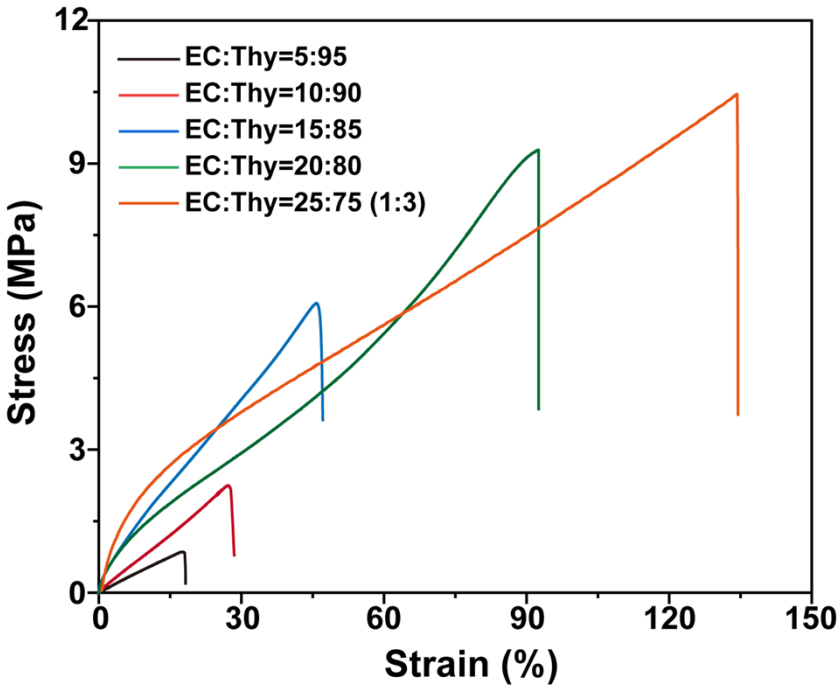

Figure S9. Stress-strain curves of CPFs prepared using EC/Thy eutectic mixtures with different ratios.

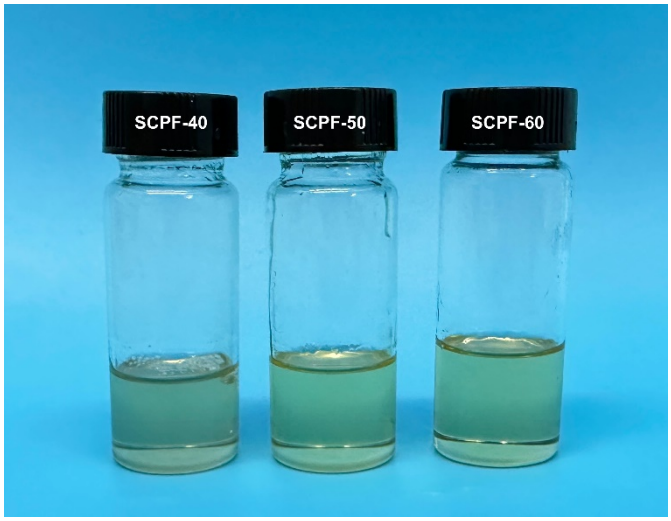

Figure S10. Optical photograph of the prepolymer solution after the introduction of stearic acid (STA), which shows good compatibility.

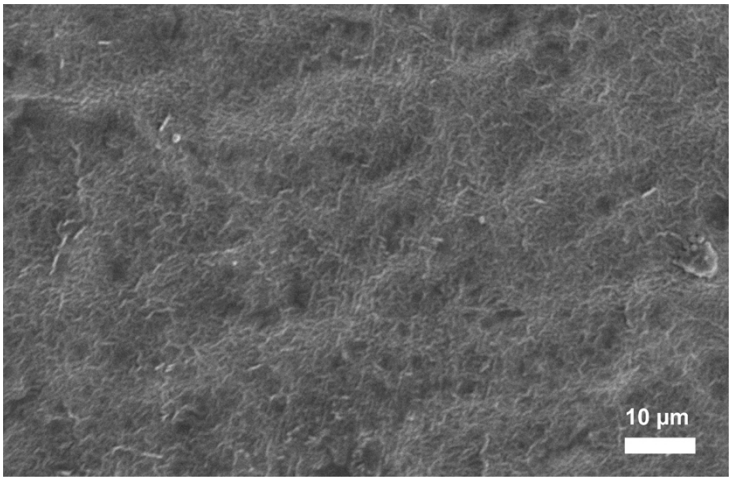

Figure S11. SEM image of the polymer obtained by direct polymerization after the introduction of STA into the prepolymer solution.

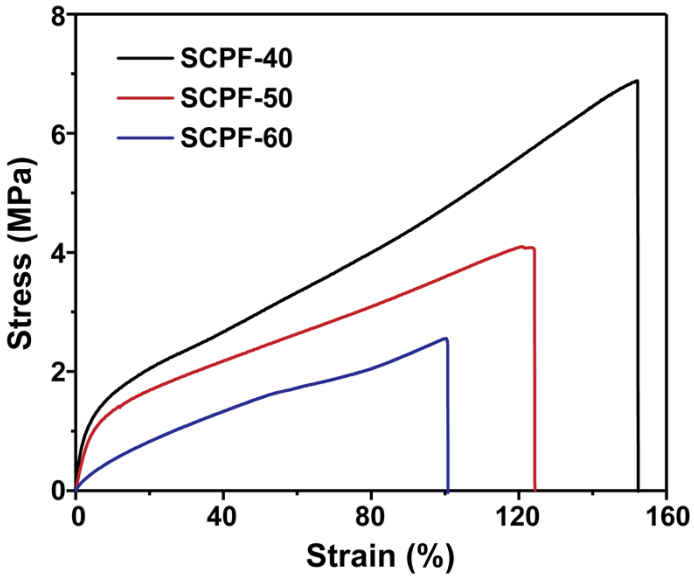

Figure S12. The stress-strain curves of different SCPFs. With the increase in STA content, the mechanical strength of SCPF decreased significantly.

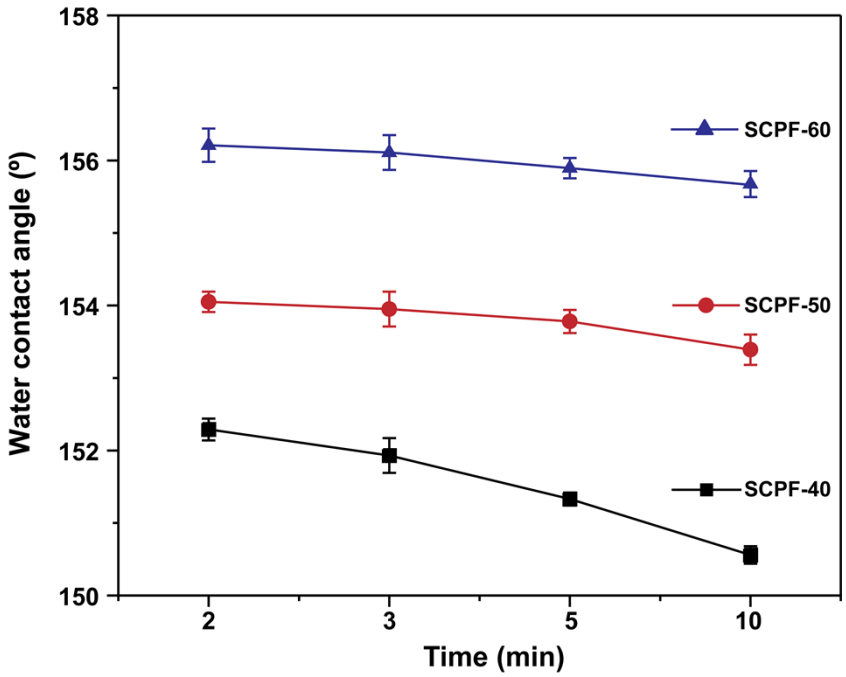

Figure S13. The change in contact angle of water droplets on the surface of SCPF after maintaining for different periods of time.

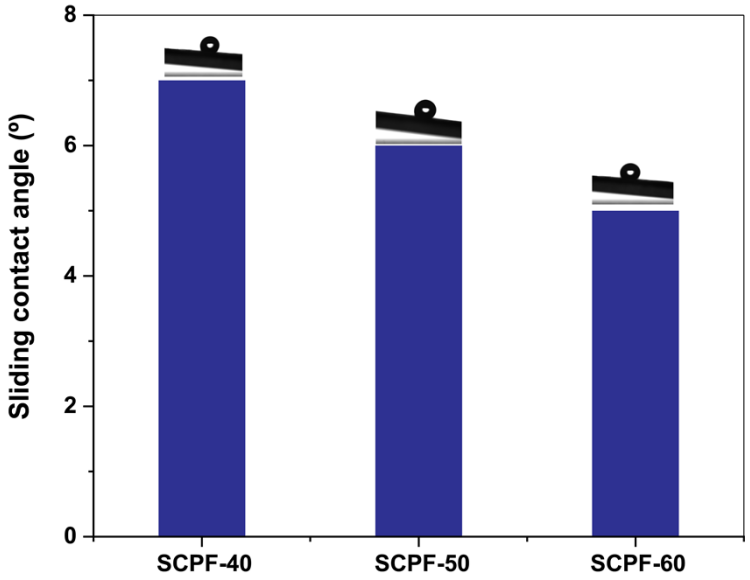

Figure S14. The sliding contact angles of different SCPFs

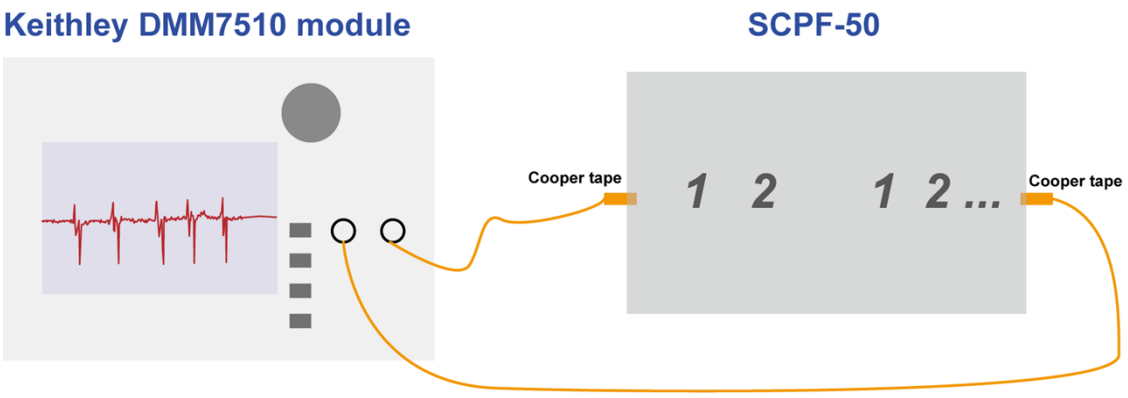

Figure S15. Schematic diagram of writing numbers on the surface of SCPF-50 and collecting electrical signals

Table S3. The component ratios used in the preparation processes of different samples

Sampe	component ratio (mass ratio)
CPF-25 (IEA)	EC:Thy:IEA =1:3:1
CPF-50 (IEA)	EC:Thy:IEA =1:3:2
CPF-75 (IEA)	EC:Thy:IEA =1:3:3
CPF-25 (EGPEA)	EC:Thy:EGPEA =1:3:1
CPF-50 (EGPEA)	EC:Thy: EGPEA =1:3:2
CPF-75 (EGPEA)	EC:Thy: EGPEA =1:3:3
CPF-15/35	EC:Thy:IEA:EGPEA =1:3:1.2:2.8
CPF-25/25	EC:Thy:IEA:EGPEA =1:3:2:2
CPF-35/15	EC:Thy:IEA:EGPEA =1:3:2.8:1.2
SCPF-40	EC:Thy:IEA:EGPEA:STA =1:3:2:2:1.6
SCPF-50	EC:Thy:IEA:EGPEA:STA =1:3:2:2:2
SCPF-60	EC:Thy:IEA:EGPEA:STA =1:3:2:2:2.4

Note: The ratio of LiTFSI and photoinitiator TPO is the same in all samples. EC:Thy:LiTFSI:TPO=1:3:0.075:0.075

Table S4. Comparison of the obtained CPF with current typical literature on preparation materials, optical transmittance, mechanical strength, hydrophobicity, and the use of solvent

Entry	Preparing materials	Optical transmittance	Mechanical strength	Hydrophobicity	The use of solvents	Ref.
1	EC, PPK	>80% at 600 nm	N/A	N/A	THF, DMF, DCM and petroleum ethe	[1]
2	EC, TEBAC, Thy, EGPEA, IBA	~92.5 %	Tensile strength:10~20 MPa Tensile strain:10~70%	Yes	DMF	[2]
3	EC, LiTFSI, Thy, Cou, EGPEA	~92%	Tensile strength:15~45 MPa Tensile strain:22~50%	Yes	DMF	[3]
4	EC, α-bromophenylaceti c acid, oxalyl chloride, LMA, FMA, DAGMA	N/A	N/A	N/A	THF, Methanol, DCM	[4]
5	EC, rosin, poly(butyl acrylate) (PBA)	40~80%	Tensile strength:1.06~14.8 MPa Tensile strain:24.8~127%	N/A	DMF, THF and petroleum ether	[5]
6	EC, NaOH, ethanol, sunflower oil	N/A	Tensile strength:0.58~13.9 MPa Tensile strain:7.5~93.9%	Yes	ethanol and water	[6]
7	EC, castor oil, Capsaicin	N/A	Tensile strength:2~5.5 MPa Tensile strain:30~60%	Yes	ethanol	[7]
8	EC, Thy, IEA, EGPEA, LiTFSI	~90%	Tensile strength:3~10.5 MPa Tensile strain:30~135%	Yes	No	This work

References

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