Supplementary Material: Strong, Tough, and Adhesive Polyampholyte/ Natural Fiber Composite Hydrogels

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S1. Supplementary Methods

S1.1. Fourier Transform Infrared Spectroscopy (FTIR)

FTIR spectra of the hydrogel samples were recorded by using a Tensor 2 FTIR spectrometer (NeXus, USA). The measured specimens were prepared by the KBr-disk method, and the spectra in the wavenumber range of 4000−400 cm^{-1} were collected with a resolution of 4 cm^{-1}. All the experiments were carried out at ambient temperature.

S1.2. Field Emission Scanning Electron Microscopy (FESEM)

Fracture morphologies of the hydrogel samples were observed by a Hitachi SU8010 FESEM. In detail, the samples were frozen and fractured in liquid nitrogen and then freeze-dried for 24 hours. Afterwards, the fractured surfaces of the samples were gold-coated in a JUC-500 Magnetron Sputtering Device (JEOL, Tokyo, Japan), and then observed with an accelerating voltage of 5 kV.

S2. Supplementary Figures

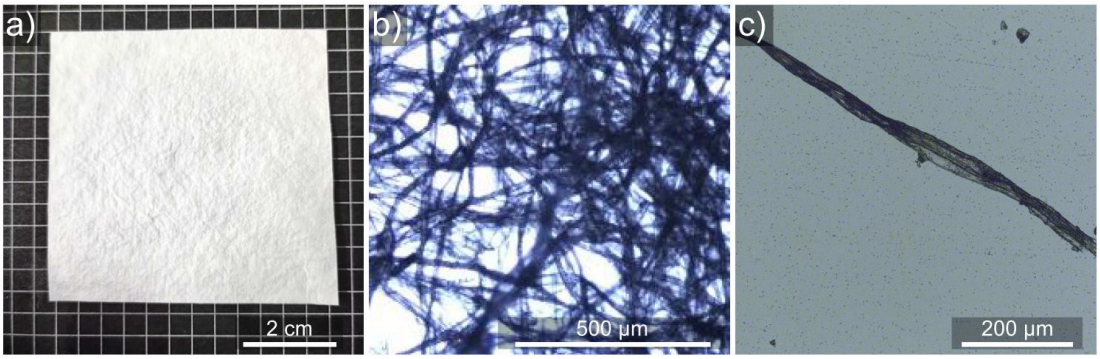

Figure S1. Macroscopic and microscopic images of cellulose-based fiber fabric (CF) sheet used in this work. (**a**) Macroscopic photograph of the dried CF sheet. (**b**,**c**) Optical microscopic photographs of the CF sheet (**b**) and a single fiber (**c**). Geometry information of the single fiber: 20−50 μm in diameter and 1.5−2 mm in length.

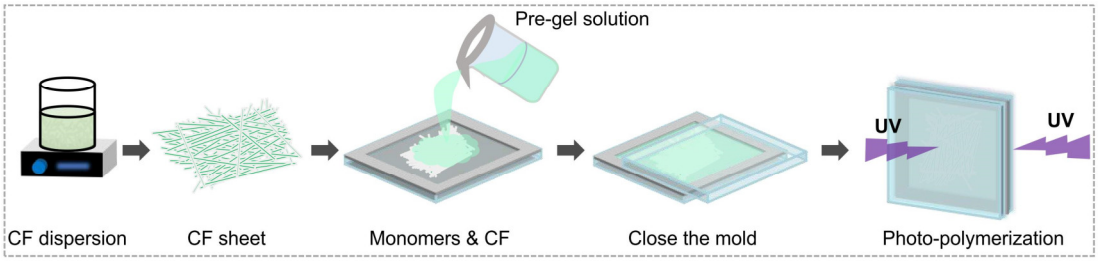

Figure S2. Fabrication process of PA/CF composite hydrogels.

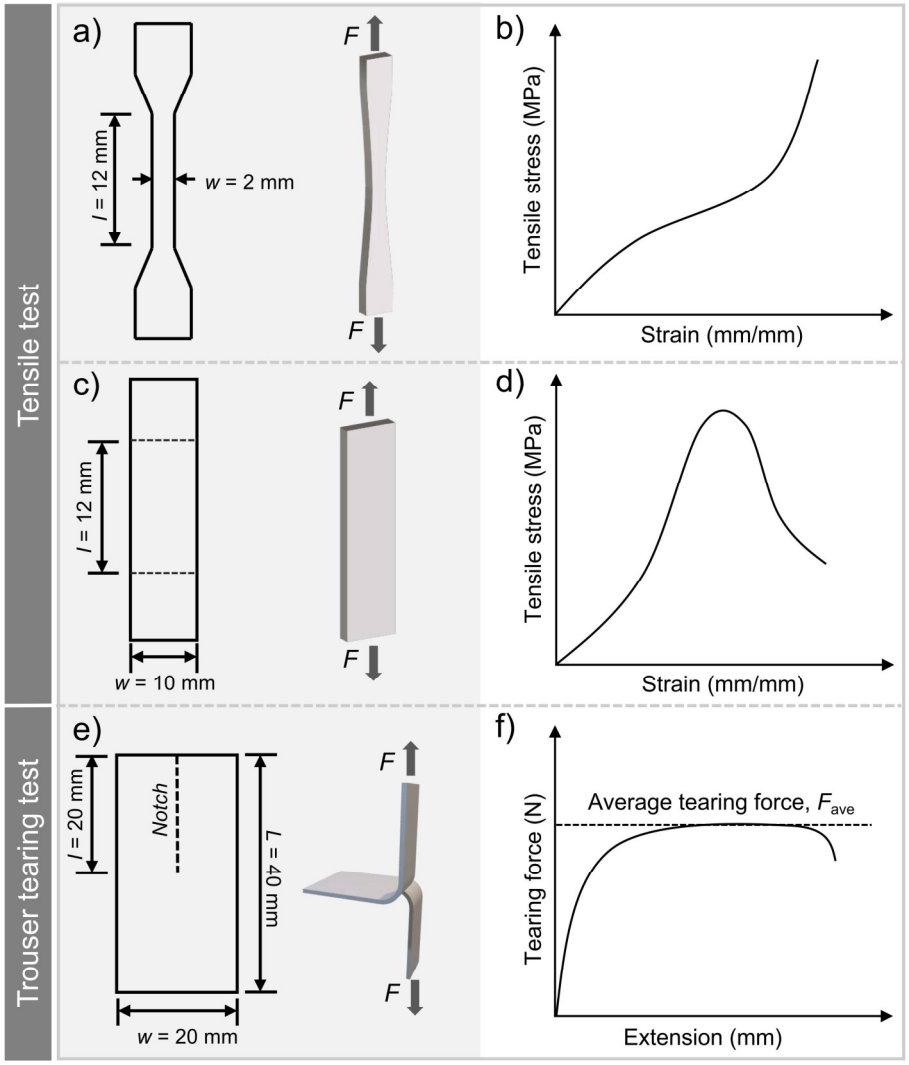

Figure S3. Sample geometries and methods for tensile and tearing tests. (**a,b**) Geometry and a representative stress-strain curve of PA gel in tensile tests. (**c,d**) Geometry and a representative stress-strain curve of PA/CF composite gels in tensile tests. (**e,f**) Geometry and a representative tearing force-extension curve of PA/CF composite gels in tearing tests. Average force (F_{ave}) obtained from the curve is used for calculation of tearing energy according to the literatures.

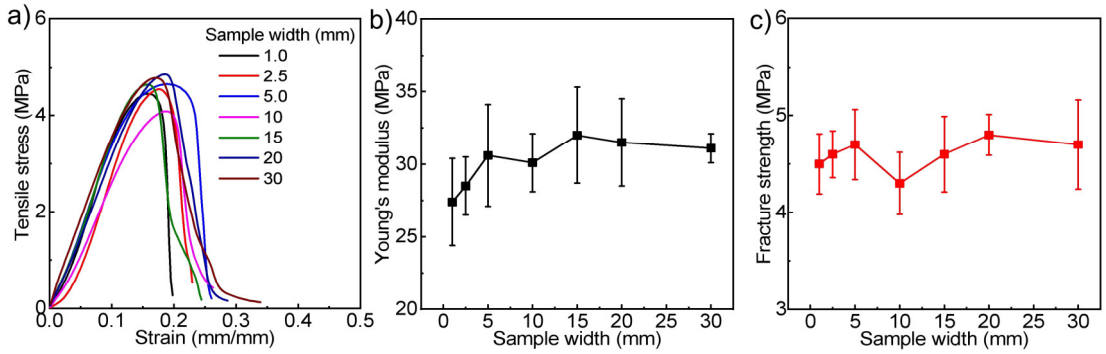

Figure S4. Mechanical properties of PA/CF composite hydrogels with different sample widths. ϕ_{CF} = 6.0 wt%. (**a**) Stress-strain curves. (**b**) Young's modulus. (**c**) Tensile fracture strength. Due to the geometrical effect, the composite gel could achieve relatively constant mechanical properties after sample width $w > 5$ mm.

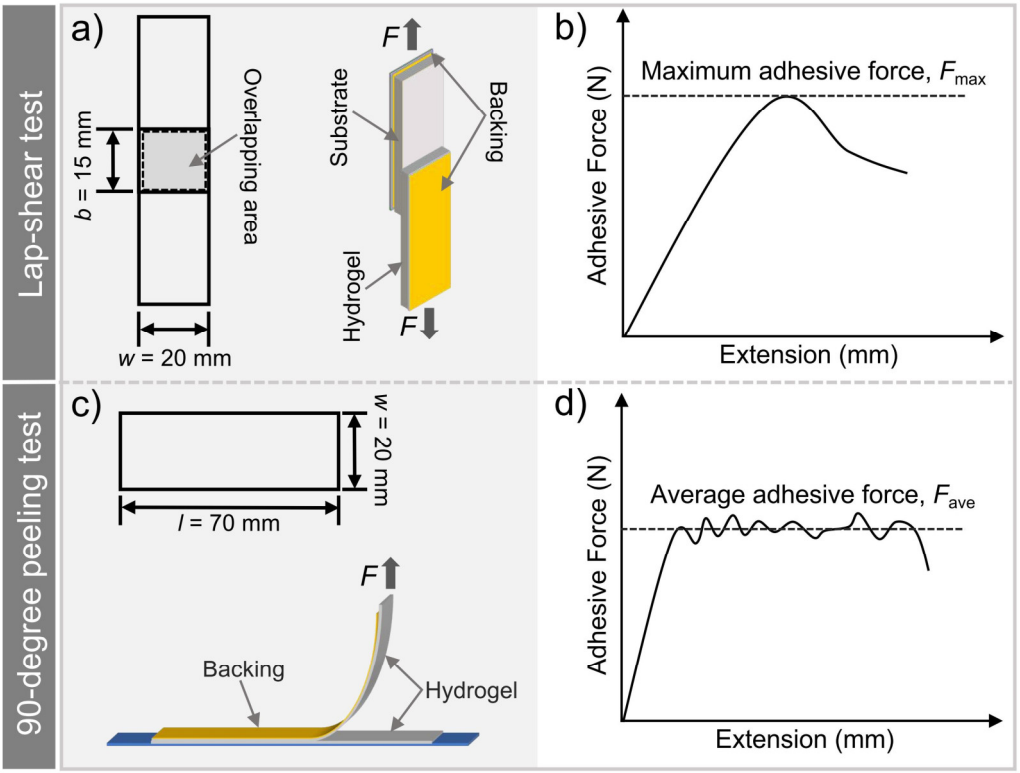

Figure S5. Sample geometries and methods for adhesion tests. (**a,b**) Geometry and a representative adhesive force-extension curve of PA/CF composite hydrogels in lap-shear tests. Maximum adhesive force (F_{max}) obtained from the curve is used for calculation of adhesive strength. (**c,d**) Geometry and a representative adhesive force-extension curve of PA/CF composite hydrogels in 90-degree peeling tests. Average adhesive force (F_{ave}) obtained from the curve is used for calculation of adhesive energy according to the literatures.

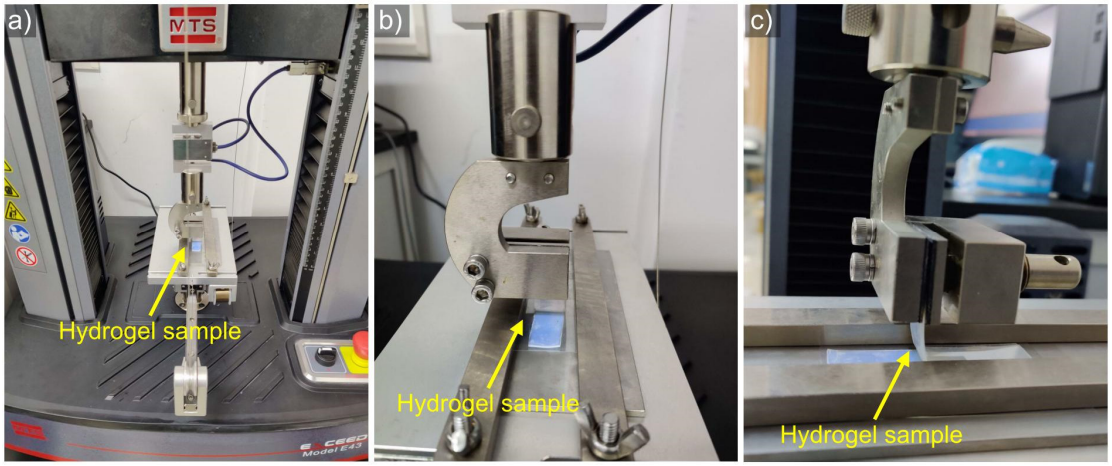

Figure S6. Photographs of 90-degree peeling tests for PA hydrogel and PA/CF composite hydrogels.

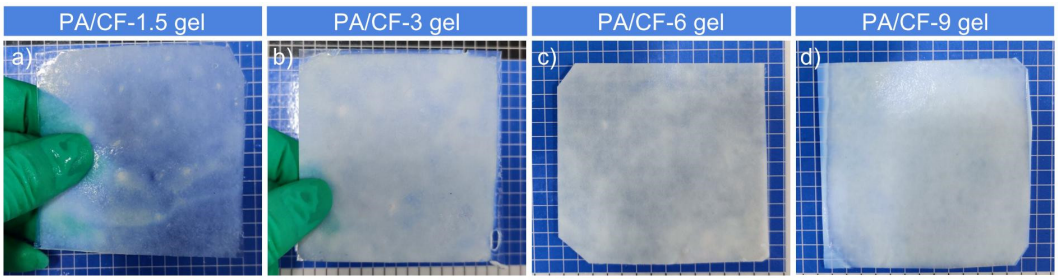

Figure S7. Macroscopic photographs of PA/CF composite hydrogels.

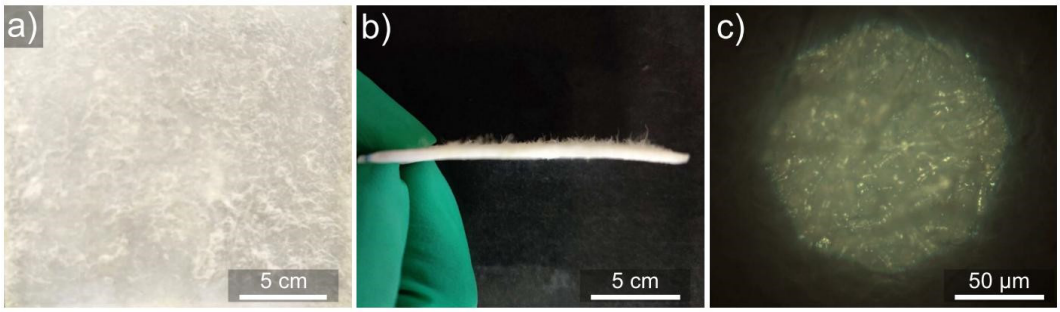

Figure S8. Failure surface of two pieces of PA/CF composite hydrogels after the lap-shear test. ϕ_{CF} = 6.0 wt%. (**a,b**) Macroscopic photographs in two directions. (**c**) Optical microscopic photograph.

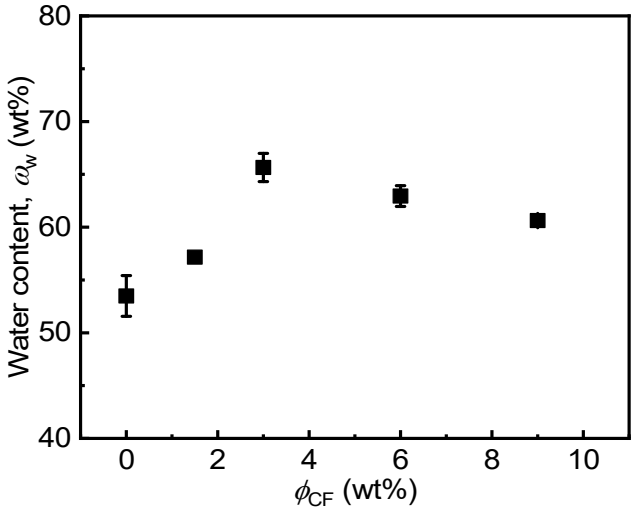

Figure S9. Water content (ω_w) versus ϕ_{CF} for PA/CF composite hydrogels.

S3. Supplementary Tables

Table S1. Summary of tensile properties of PA/CF composite hydrogels with different sample widths (shown in **Figure S4**).

Sample width (mm)	E (MPa)	σ_b (MPa)	W_b (MJ m^{-3})
1.0	27.4 ± 3.0	4.5 ± 0.3	0.53 ± 0.05
2.5	28.5 ± 2.0	4.6 ± 0.2	0.67 ± 0.06
5.0	30.6 ±3.5	4.7 ± 0.4	0.79 ± 0.07
10	30.1 ± 2.0	4.3 ± 0.3	0.58 ± 0.04
15	32.0 ± 3.3	4.6 ± 0.4	0.60 ± 0.03
20	31.5 ± 3.0	4.8 ± 0.2	0.73 ± 0.05
30	31.1 ± 1.0	4.7 ± 0.4	0.69 ± 0.06

Table S2. Summary of tensile properties of representative hydrogel samples (shown in **Figure 1c,d,e**).

Sample code (ϕ) [a)]	E (MPa)	σ_b (MPa)	W_b (MJ m^{-3})
Neat PA gel	1.7 ± 0.3	2.0 ± 0.2	5.9 ± 0.4
PA/CF-ϕ gel	29.3 ± 1.7	4.3 ± 0.4	0.6 ± 0.1
Neat CF sheet	10.4 ± 0.9	0.3 ± 0.1	0.01 ± 0.01

[a)] ϕ in the code of PA/CF-ϕ represents weight percentage of CF in the corresponding PA/CF composite hydrogel.

Table S3. Summary of adhesion strength of PA/CF-6 composite hydrogels to diverse substrates (shown in **Figure 8**).

Substrates	PA/CF-6 hydrogel	Glass	Cu	PET
τ_s (kPa)	940 ± 120	404 ± 60	215 ± 20	25 ± 5