SUPPLEMENTARY DATA

Mechanically Strong Fully Biobased Anisotropic Cellulose Aerogels

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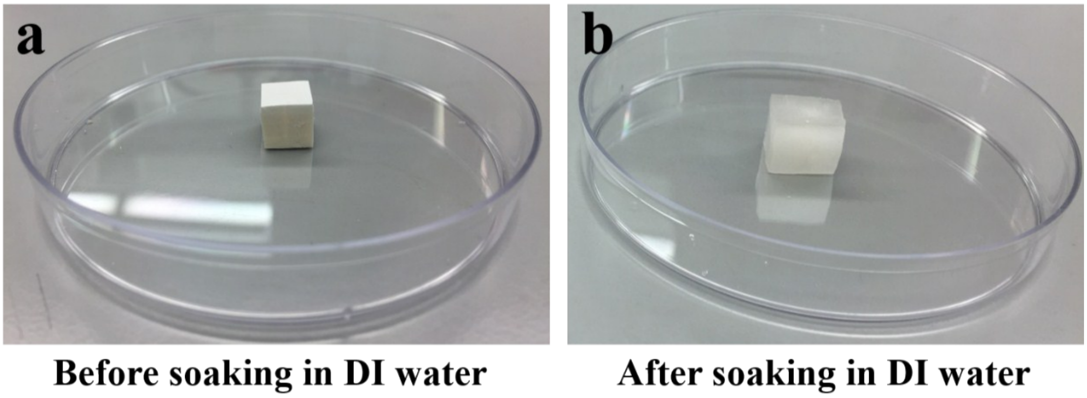

Fig. S1 Sample HD-CNF-5 before being soaked in the DI water (a) and after being soaked in DI water for 72 h (b).

Silanization of the CMC/CNF Aerogels

Silanization of the CMC/CNF aerogels was confirmed using FTIR analysis (Fig. S2). A peak at 1730 cm^{-1} was observed which could be attributed to carbonyl ester stretching vibrations in COO-Si units. The absorption bands at 780 cm^{-1} and 1271 cm^{-1} were ascribed to the characteristic vibrations of Si-O-Si and C-Si asymmetric stretching in C-Si-O units, respectively.

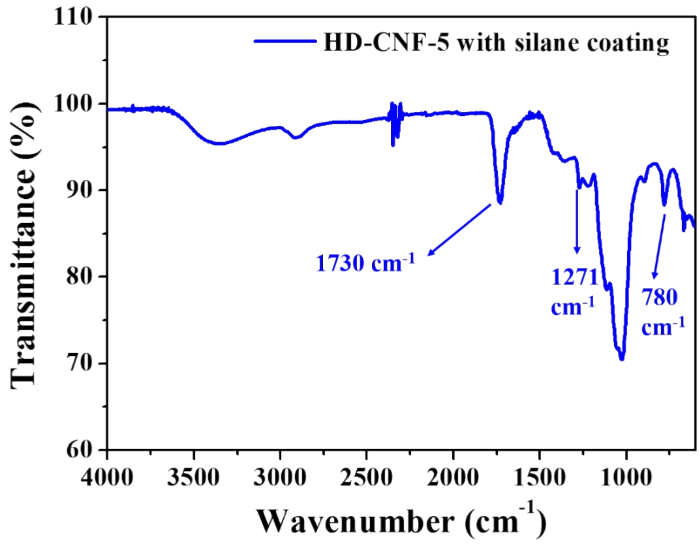

Fig. S2 FTIR spectrum of Sample HD-CNF-5 with the silane coating.

Surface Wettability of the Aerogels

Cellulose aerogels are hydrophilic and could absorb moisture in the air, which would limit their applications. To obtain hydrophobic aerogels, the CMC/CNF aerogels were treated with methyltrichlorosilane *via* a simple thermal chemical vapor deposition process. The surface wettability of the CMC/CNF aerogel was investigated using a contact angle measurement. The average contact angle and standard deviation were shown in Fig. S3. For the uncoated CMC/CNF aerogels, water droplets were completely absorbed within 10s (Fig. S3 (a) and (b)). However, the silane-coated CMC/CNF aerogel become hydrophobic (Fig. S3 (c)). The contact angles of sample HD-CNF-0, HD-CNF-5 and LD-CNF-5 were 122.1 ± 6.5°, 103.0 ± 1.2° and 114.5 ± 8.5° after the water droplet was deposited on the surface for 120s, respectively (Fig. S3 (d)). The hydrophobic aerogels can be potentially used as the mechanically strong building materials.

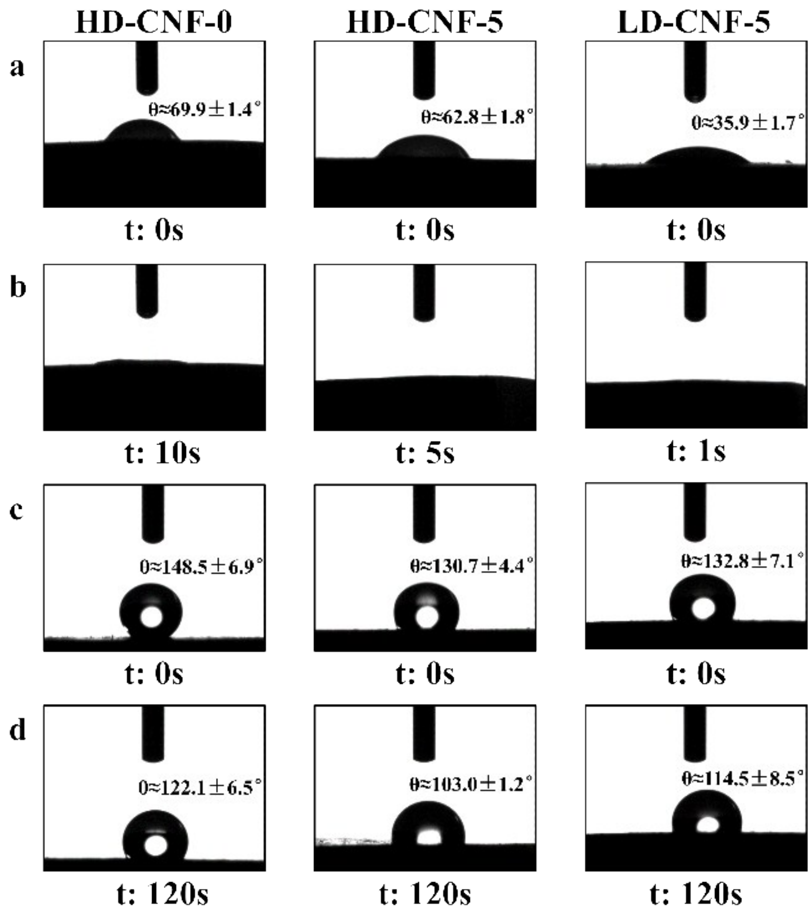

Fig. S3 Water contact angle measurements of the CMC/CNF aerogels: (a) and (b) uncoated aerogel; (c) and (d) silane-coated aerogel.

Compressive Modulus of the Non-crosslinked/Crosslinked CMC/CNF Aerogels

As shown in Fig. S4, the moduli of crosslinked aerogels were only slightly higher than those of non-crosslinked aerogels. For instance, the modulus of sample *v*-HD-CNF-5-crosslinked was 10.7 MPa, while the modulus of sample *v*-HD-CNF-5-non-crosslinked was 10.1 MPa. These results demonstrated the crosslinking only make a minor contribution to improving the mechanical performance of the aerogels. These results suggested the dramatic improvement of the mechanical properties of the CMC/CNF aerogels were attributed to the intrinsic excellent mechanical properties of

CNFs and the anisotropic structure of the aerogels.

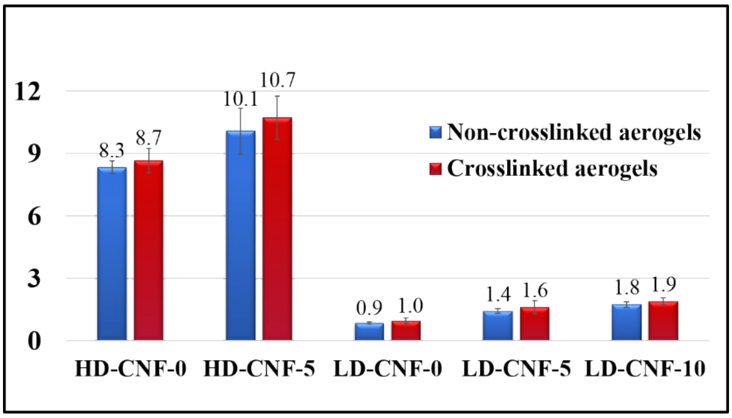

Fig. S4 Comparison of the compressive moduli of the non-crosslinked and crosslinked CMC/CNF aerogels in the vertical direction.

Heavy Metal Ion Adsorption Capacity Measurements

Solutions of heavy metal ions including Ag^{+}, Cu^{2+}, Pb^{2+}, and Hg^{2+} were prepared by dissolving the required quantity of salts in the DI water under stirring at room temperature for 3 h. The aerogel sample (30 mg) was soaked in the heavy metal solution (50 ml, 50 mg/L) under mild stirring at room temperature for 3 days. The concentration of the heavy metal ion remaining in the solution was determined using an inductively coupled plasma (ICP) atomic emission spectrometer (Optima 2000, PerkinElmer Inc., USA). The heavy metal ion adsorption capacity (q (mg g^{-1})) of the aerogels was calculated following Equation (S1),

$$q = \frac{(C_0 - C_{eq}) \times V}{M} \tag{S1}$$

where C_0 and C_{eq} (mg/L) are the initial and equilibrium concentrations of metal ions in the solution, respectively; V (L) is the volume of the metal ion solution, and M (g) is the weight of the aerogel. At least 3 samples were measured for each of the metal ions. The average results and the standard deviation are reported.

Heavy Metal Ion Adsorption of the Various Aerogels

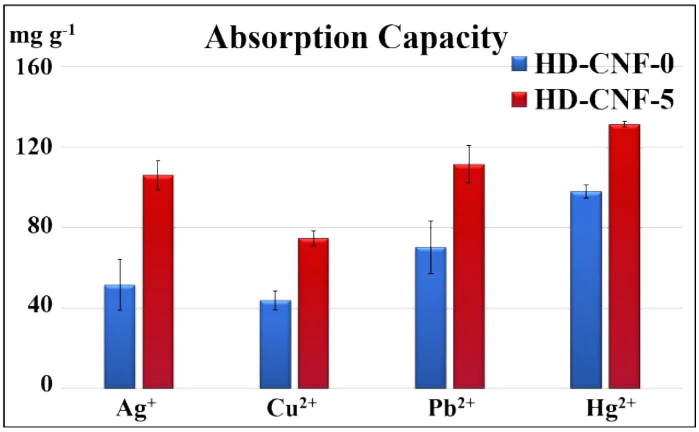

Fig. S5 Heavy metal ion adsorption capabilities of the HD-CNF-0 and HD-CNF-5 aerogels.

Table S1 The mechanical properties of some aerogels in the literature

Aerogel Materials	Density g cm^{-3}	Compressive Modulus MPa	Flexural Modulus MPa	Reference
CMC/CNF aerogel	0.105	10.7	54.9	Our study
CMC aerogel	0.12	3.4	—	1
Nanofibrillated cellulose aerogel	0.105	2.8	—	2
Porous cellulose material	0.1	2.4	—	3
Pectin aerogel	0.115	5	—	4
Polyimide aerogel	0.194	11.1	—	5
Methyltrimethoxysilane based aerogel	0.105	0.5	—	6
Polymer crosslinked silica aeroge	0.106	—	1.2	7
Silica aerogel	0.13	1.15	1.47	8
Amine-modified silica aerogels	0.11	—	0.29	9

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