Supporting Information

Highly Compressible, Anisotropic Aerogel with Aligned Cellulose Nanofibers

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The supplementary information file includes 12 figures (Figure S1- S12) and 1 table (Table S1).

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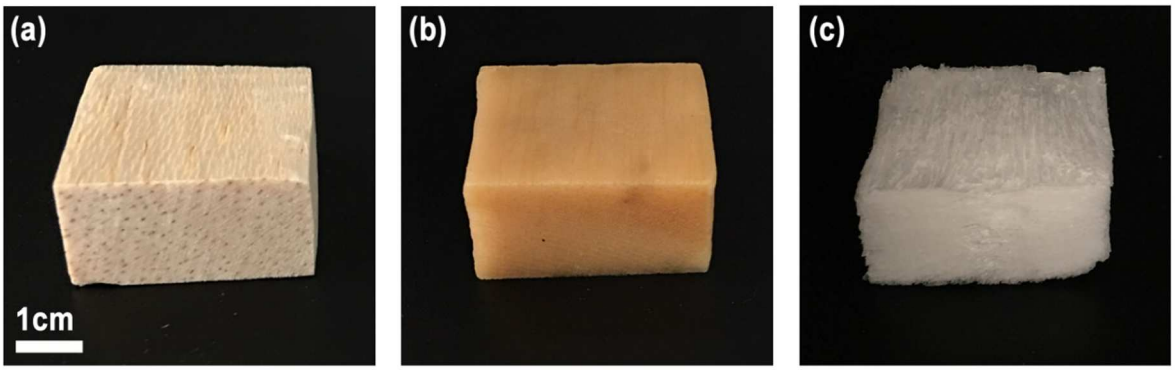

Figure S1. Photo images showing the color evolution of the wood samples by chemical treatment: (a) natural wood, (b) wood sample after $NaOH/Na_2SO_3$ treatment, (c) wood aerogel resulted from further $NaOH/Na_2SO_3$ and H_2O_2 treatment.

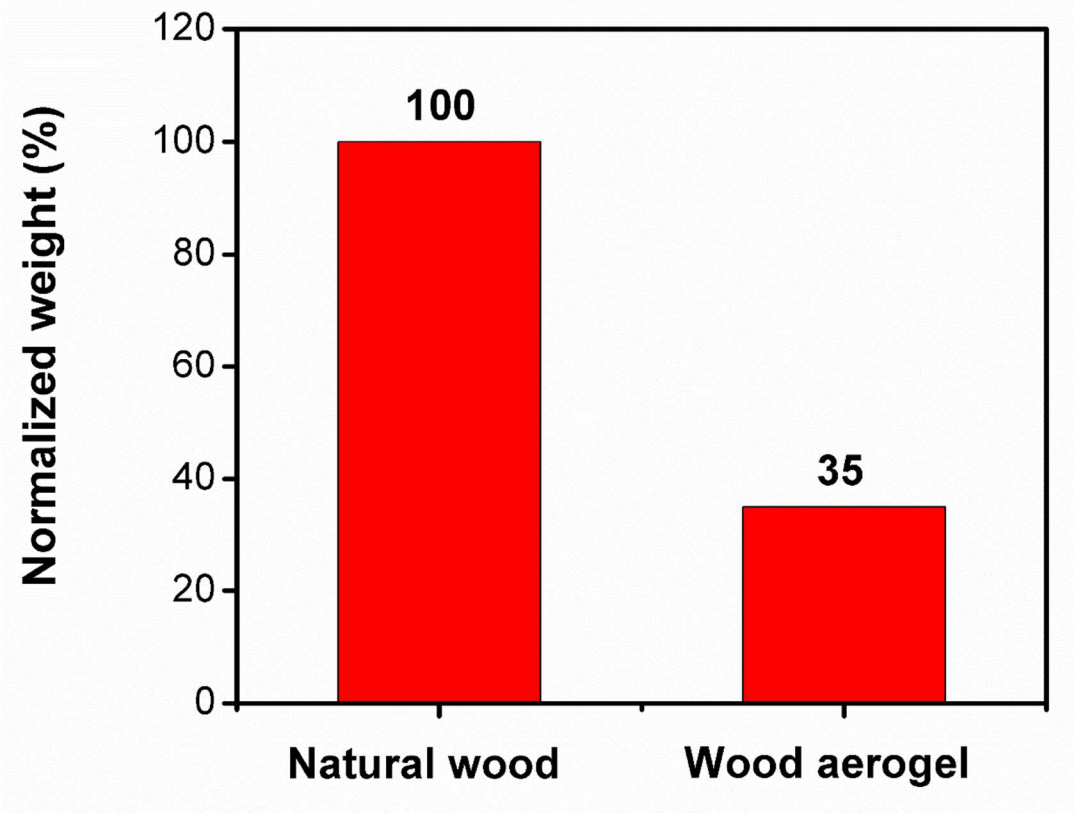

Figure S2. Weight loss of the wood samples before and after chemical treatment.

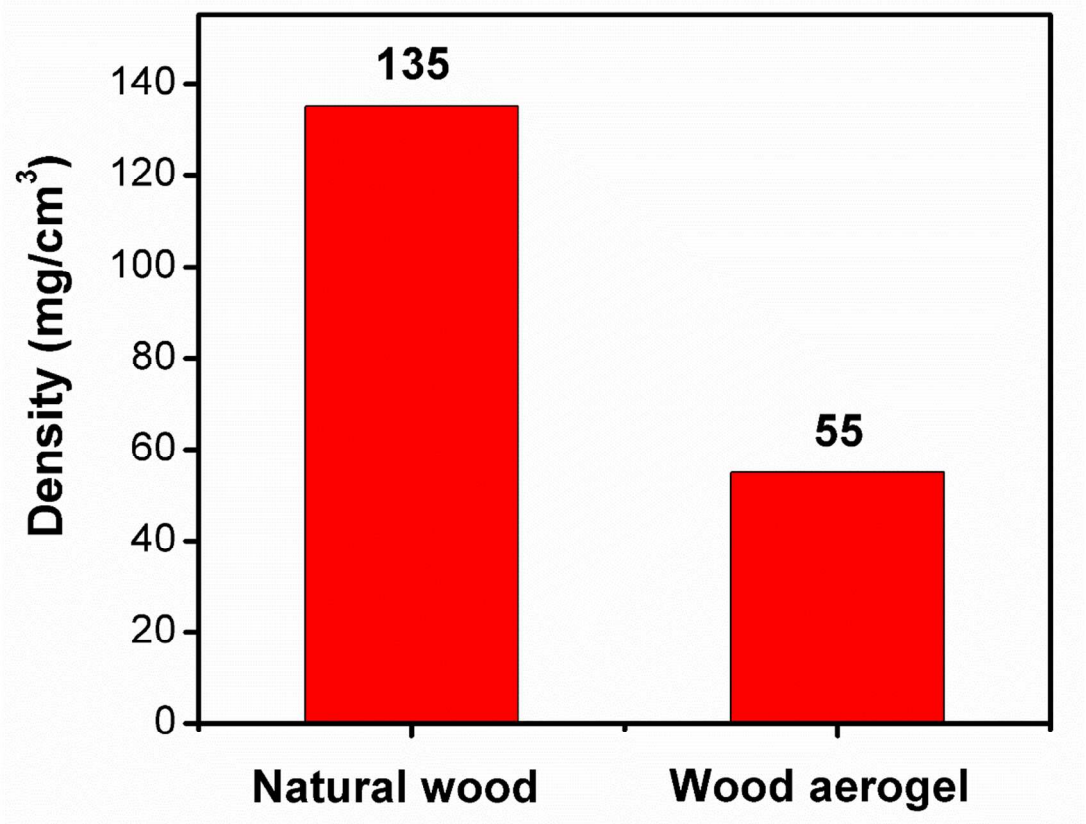

Figure S3. Density comparison from natural wood to wood aerogel, showing the significant decrease of density after chemical treatment.

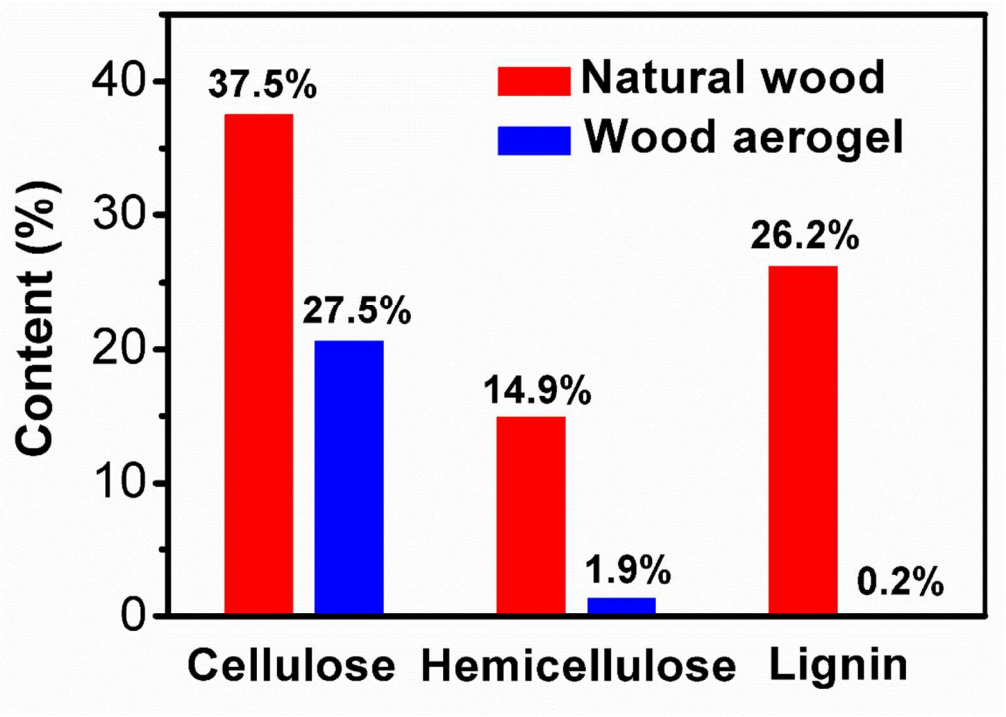

Figure S4. The cellulose, hemicellulose and lignin content evolution from natural wood to wood aerogel.

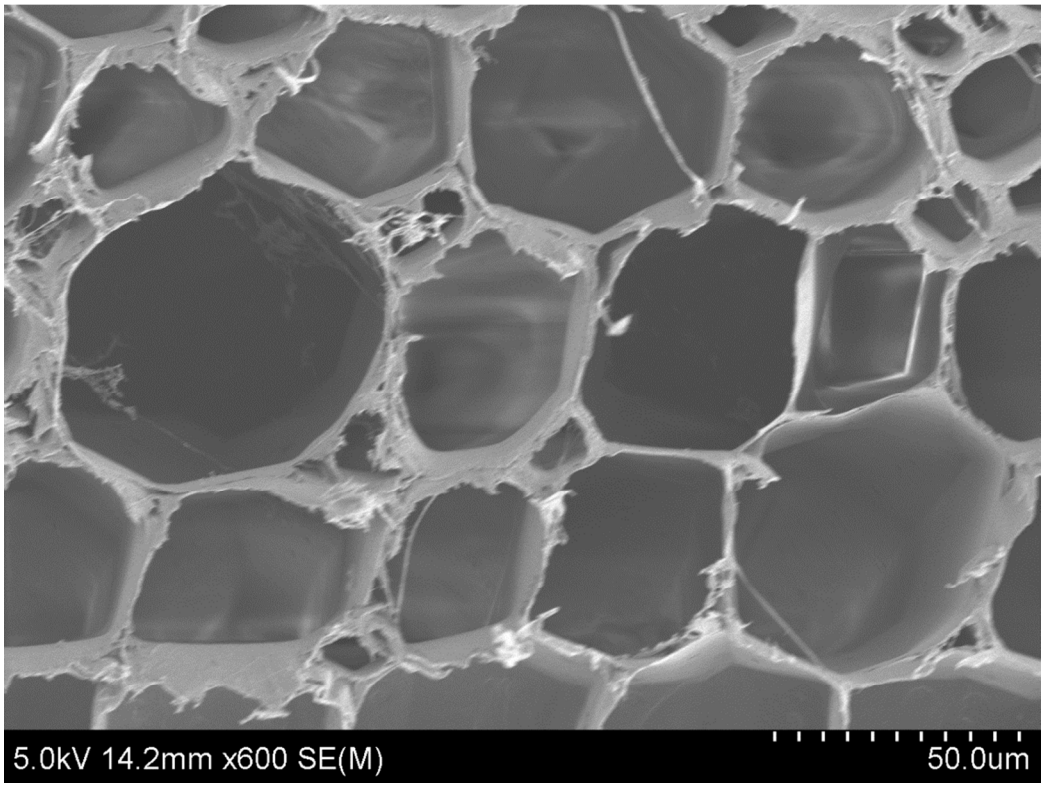

Figure S5. SEM image of natural wood, showing the wood lumen structure with thin cell wall.

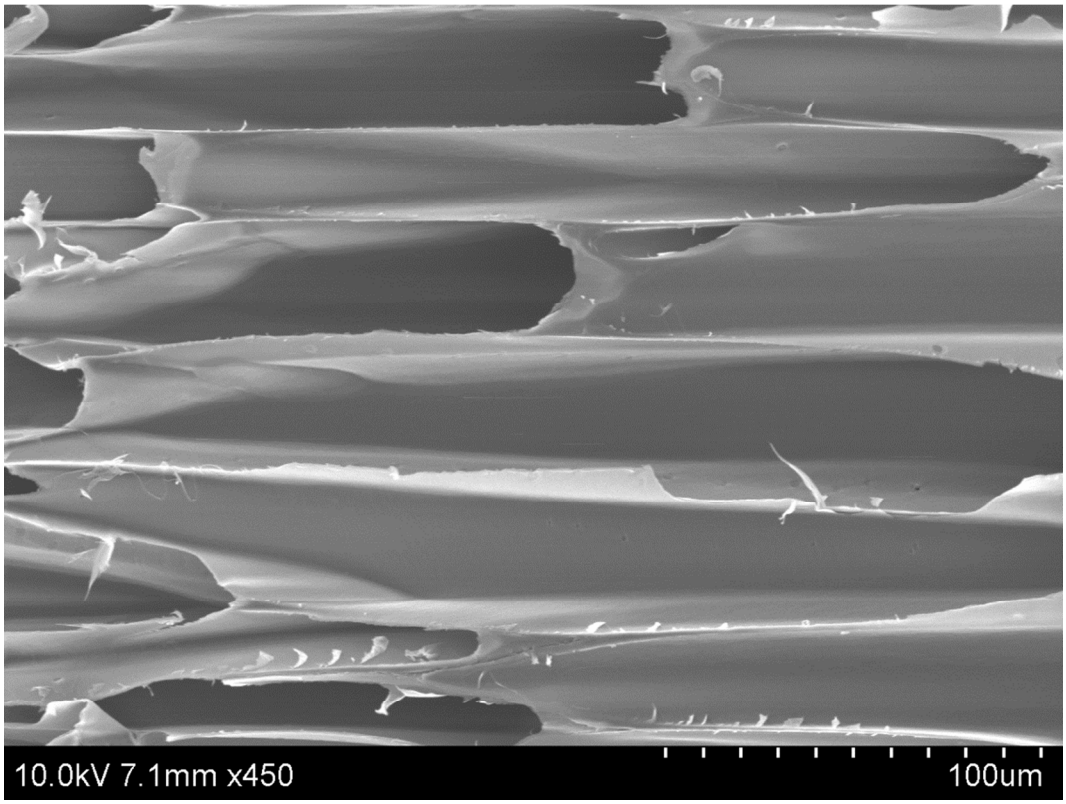

Figure S6. SEM image of natural wood, showing the lumina along the tree-growth direction.

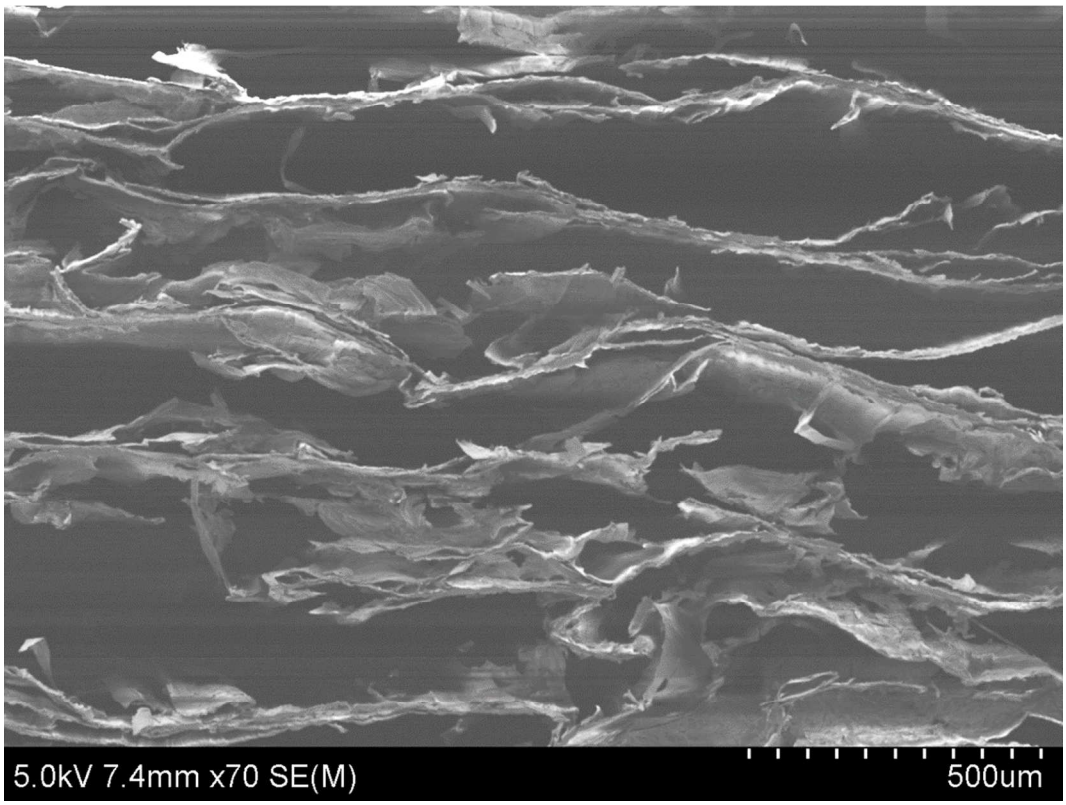

Figure S7. SEM image of wood aerogel, showing that the wood lumina become layered structure after chemical treatment.

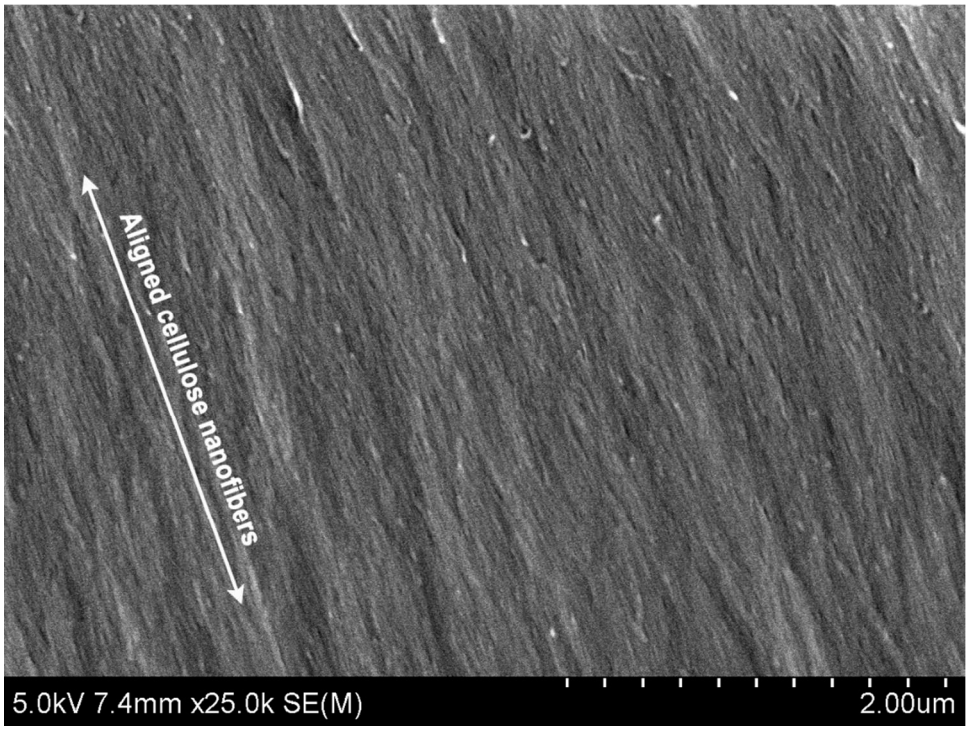

Figure S8. Magnified SEM image of the wood aerogel, showing good alignment of cellulose nanofibers.

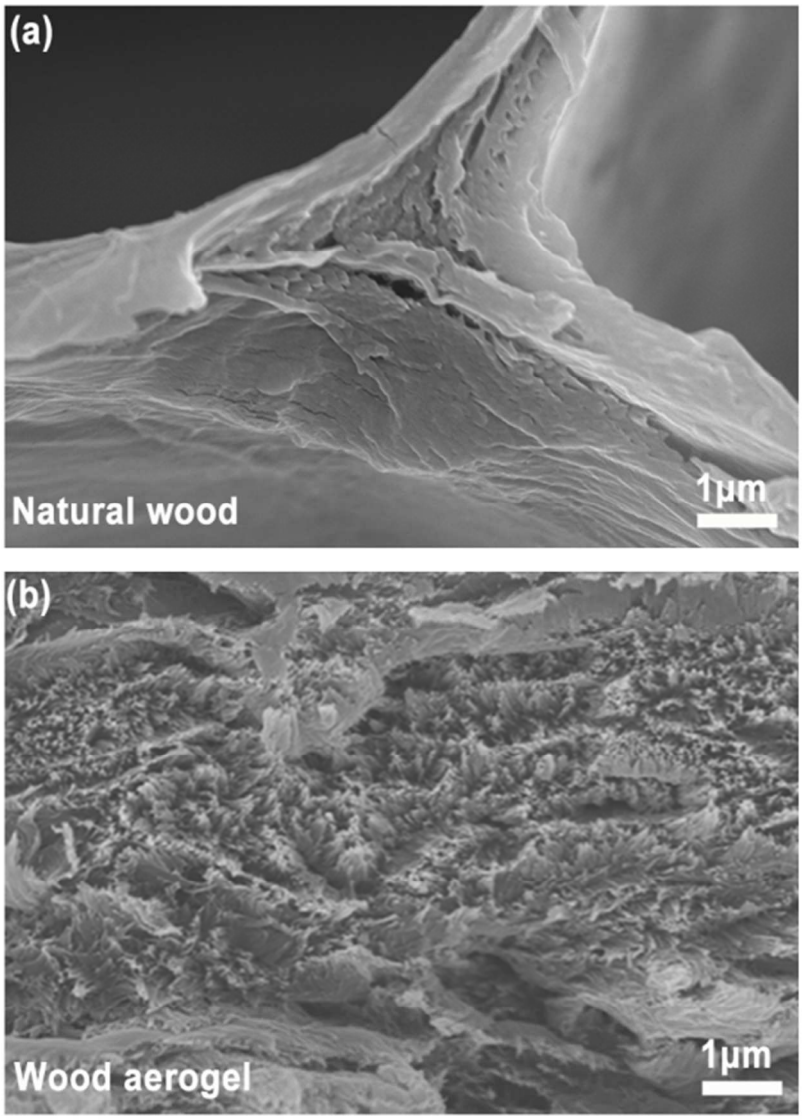

Figure S9. Top-view magnified SEM images of natural wood (a) and wood aerogel (b), showing the wood lumen structure change from dense cell wall to highly porous cell wall with isolated cellulose nanofibers after chemical treatment due to the removal of lignin and hemicellulose.

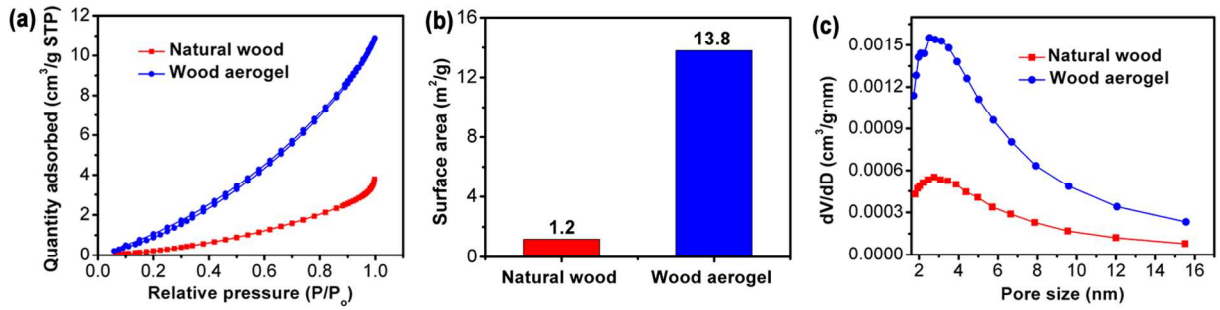

Figure S10. (a) Nitrogen adsorption-desorption isotherms of natural wood and wood aerogel. (b) Surface area comparison from natural wood to wood aerogel. (c) Pore size distribution of natural wood and wood aerogel, indicating the generation of nanopores due to hemicellulose/lignin removal.

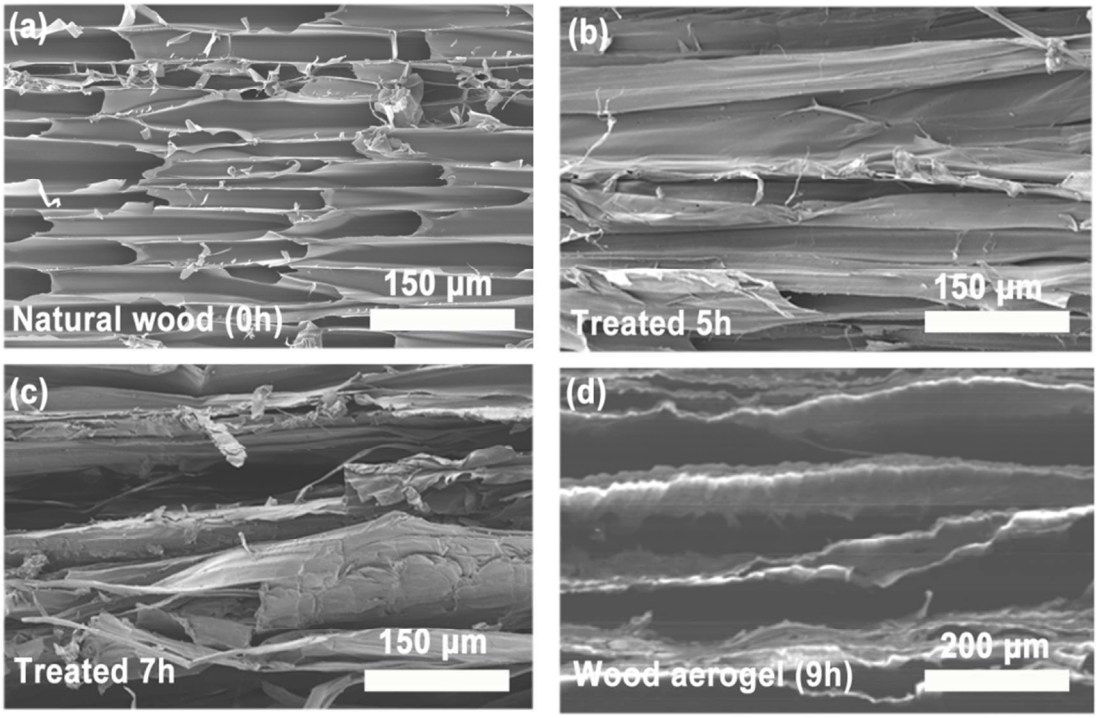

Figure S11. SEM images showing the wood lumen structure evolution with different chemical treating time: (a) natural wood (0h), (b) after 5h-treatment, (c) after 7h-treatment, (d) wood aerogel (9h).

The treated wood sample were prepared by cooking the dry natural wood in a mixture solution of Sodium Hydroxide (NaOH) and Sodium Sulfite (Na_2SO_3) at 100 □ for 5h, followed by boiling in the H_2O_2 solution to complete remove the residual lignin. The yield (Y) was calculated as:

$$Y = [m_i/m_0]*100\% \qquad (1)$$

Where m_i is the weight of treated wood sample, m_0 is the weight of dry natural wood sample.

The cellulose, hemicellulose, and lignin contents were measured by the standard protocol used at the USDA Forest Products Lab (*Ind. Eng. Chem. Res., 2010, 49, 8258-8266*) to get the test value of each sample. As each sample has different yield, we normalized the test values of all samples to compare the evolutions of the three components in all samples. The normalized value of

content for cellulose, hemicellulose and lignin were calculated as:

$$C_{(normalization)} = [C_{(test)*}Y]*100\% \qquad (2)$$

where $C_{(test)}$ is the test value of cellulose (or hemicellulose, or lignin) content, Y is the yield, and C(normalization) is the normalized value of cellulose (or hemicellulose, or lignin) content. All the test and normalized values of cellulose, hemicellulose, and lignin for the four samples are summarized in Table S1.

Table S1. The chemical composition evolution from natural wood to wood aerogel.

		Cellulose (%)	**Hemicellulose (%)**	**Lignin (%)**
Natural wood (Treated 0h)	Test value (After normalization)[a]	37.5	14.9	26.2
Treated 5h	Test value	63.0	6.0	21.4
	Yield	57.0%		
	After normalization	35.9 (63.0*57%)	3.4 (6.0*57%)	12.2 (21.4*57%)
Treated 7h	Test value	73.8	2.9	3.5
	Yield	44.6%		
	After normalization	32.9 (73.8*44.6%)	1.3 (2.9*44.6%)	1.6 (3.5*44.6%)
Wood aerogel (Treated 9h)	Test value	78.5	5.4	0.6
	Yield	35.0%		
	After normalization	27.5 (78.5*35%)	1.9 (5.4*35%)	0.2 (0.6*35%)

Note:

a. As the yield value of natural wood is 100%, the normalization value is equal to the test value.

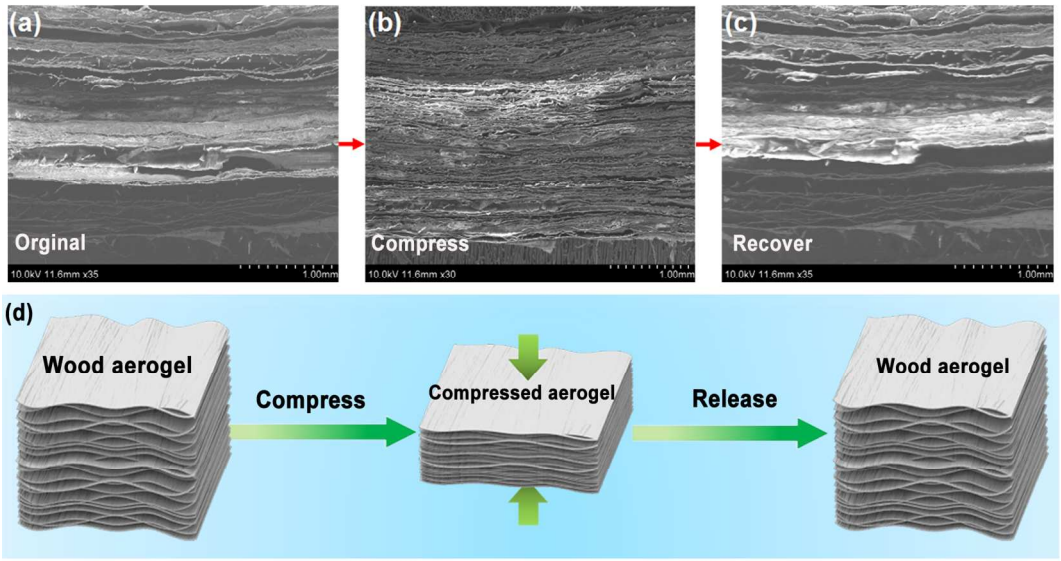

Figure S12. (a-c) Ex-situ SEM images of the wood aerogel: (a) before compressing, (b) under compressing (c) after recovery. (d) Graphical illustration of the wood aerogel before compressing, under compressing and after release, respectively.

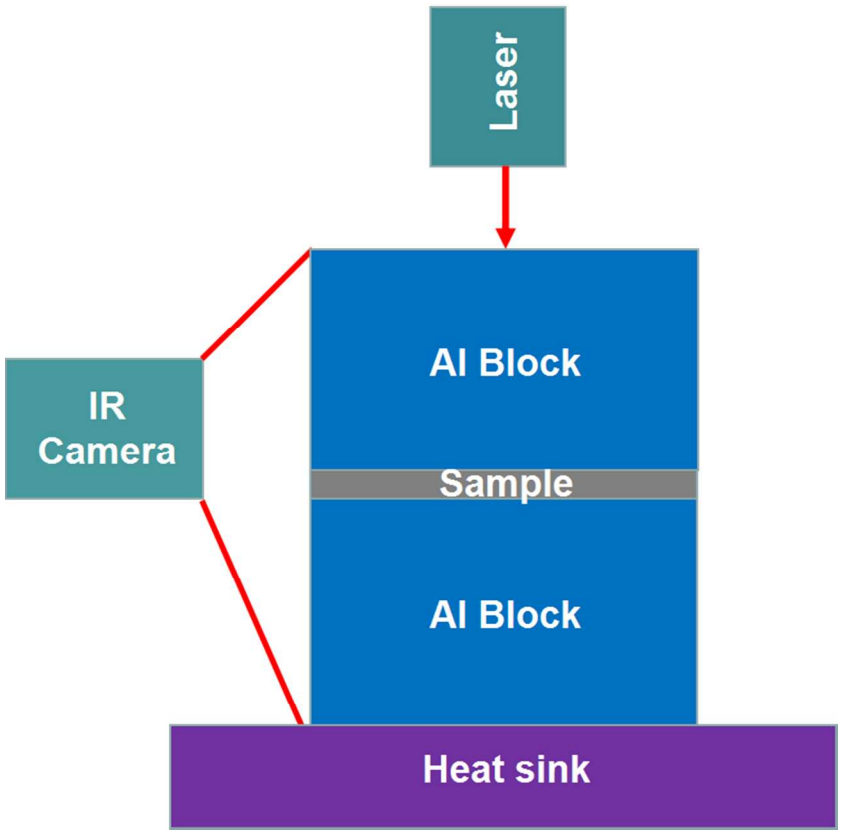

Figure S13. Schematic illustration of the set up for thermal conductivity measurement.