Supporting Information

Anisotropic, Ultrastrong and Light-transmission film Designed on Wheat Straw

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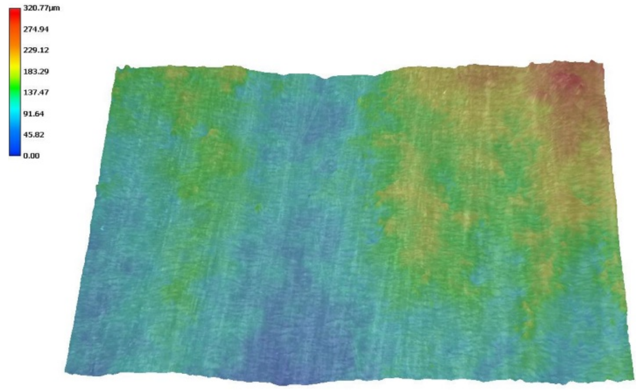

Figure S1 Image of the wheat straw film. The flat surface of WSF observed under ultra-deep field microscope.

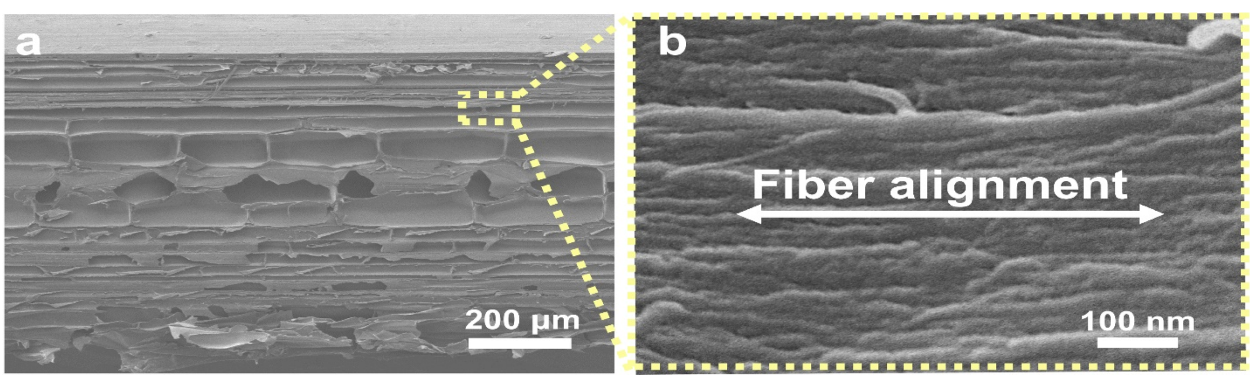

Figure S2. SEM of the longitudinal section of the OWS.

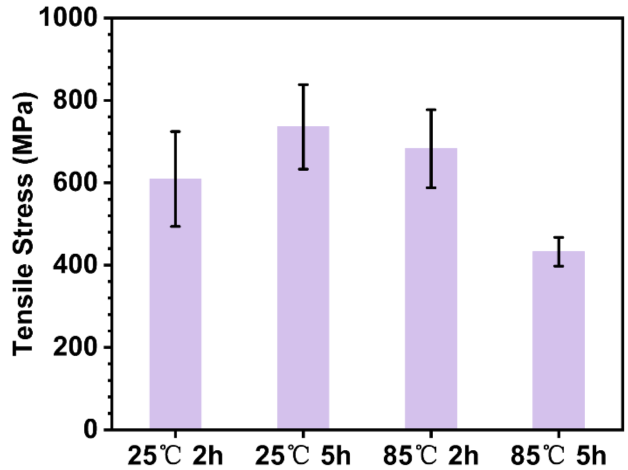

Figure S3. Effect of different ways of sodium hydroxide pretreatment on the mechanical properties of the WSF. Excessive temperature and time led to the destruction of the amorphous region in the cellulose straw.

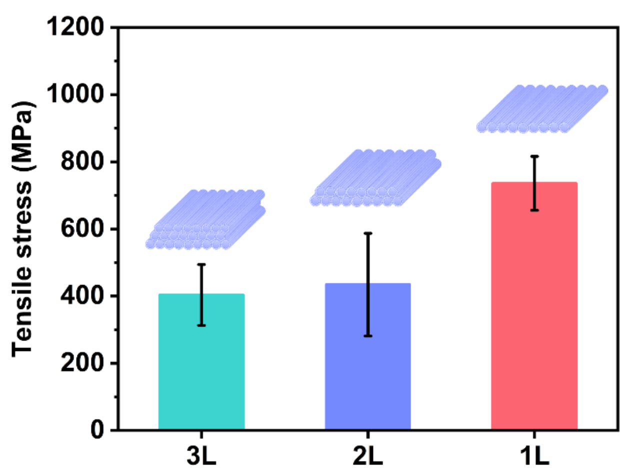

Figure S4. Tensile strength of WSF decreased with an increasing number of DWS layers. This might be due to the weak bonding between the layers. Although the tensile stress of two- and three-layer WSF film decreased, it was still higher than that reported in many other similar studies.[1, 2]

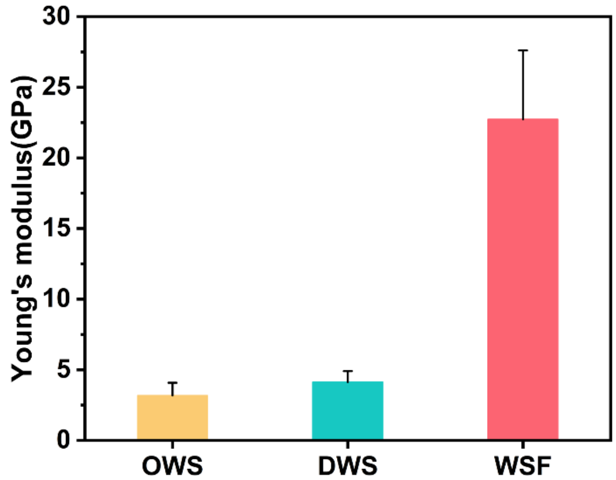

Figure S5. Young's modulus of OWS, DWS, and WSF.

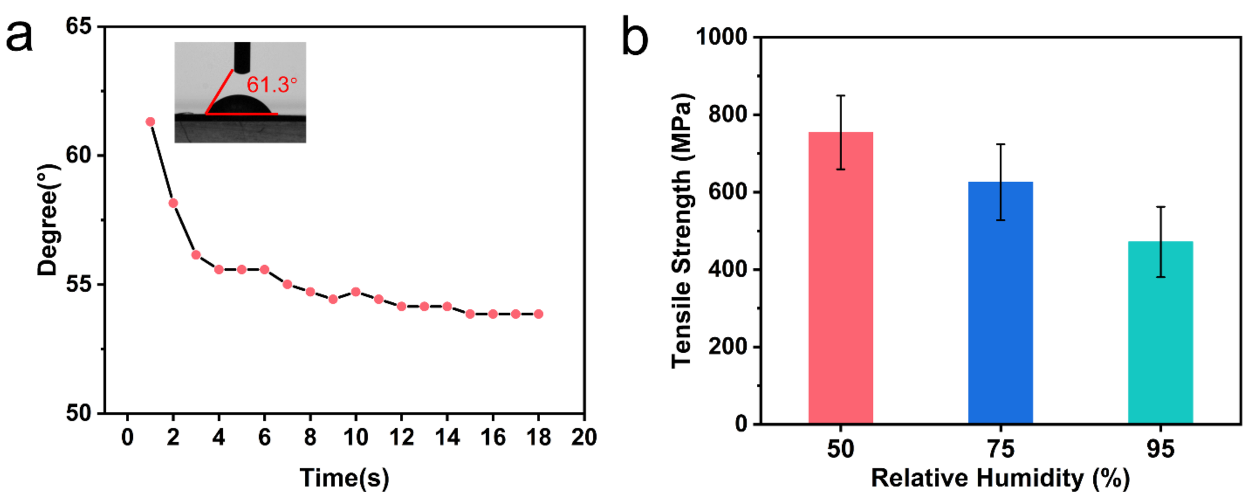

Figure S6. (a) Water contact angle and dynamic water absorption of WSF. (b). WSF tensile strength at different relative humidity.

The initial water contact angle was 61.3°, which decreased to 53.8° after 18 s (Fig. S10a). Similarly, the tensile strength of WSF decreased with increasing relative humidity (RH) when WSF was subjected to 50%, 75%, and 95% RH for 24 h. These phenomena could be attributed to the hydrophilic nature of cellulose.

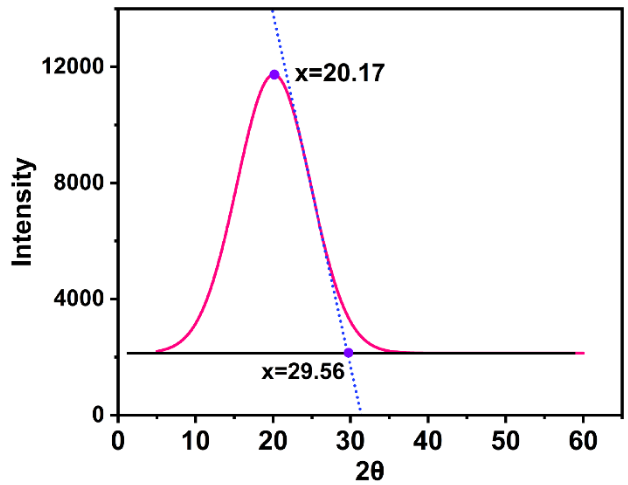

Figure S7. Gaussian fitted curve of the OWS.

The microfibril angles were determined by Cave's method in combination with XRD. The parameter T was used as an indicator of the angle, and the straw microfibril angle (MFA) was obtained using Cave's equation:[3] MFA = 0.6T. The inflection point on the intensity distribution curve was found, a graphical tangent line was drawn through the inflection point, and half of the distance between the two points intersecting the baseline was taken as the angular distance T. MFA = 0.6 ×(29.56 – 20.17) = 5.634°.

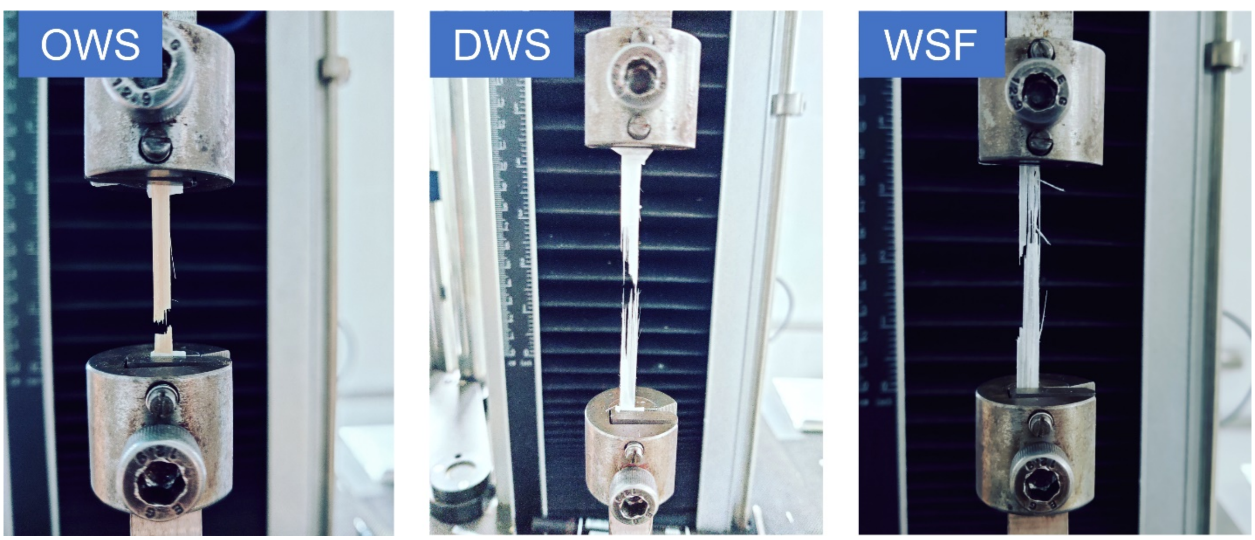

Figure S8. Photograph of the tensile fracture of OWS, DWS, and WSF.

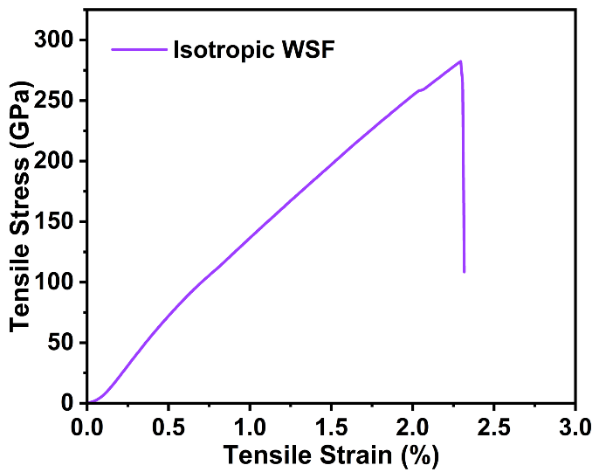

Figure S9. Typical strain-stress curves of Isotropic WSF.

The isotropic WSF was prepared by the cross-arrangement of the two layers of DWS. Its tensile strength was 302.83 ± 48 Mpa, which was higher than the previously reported strength of cellulose film (≈258 MPa). [4]

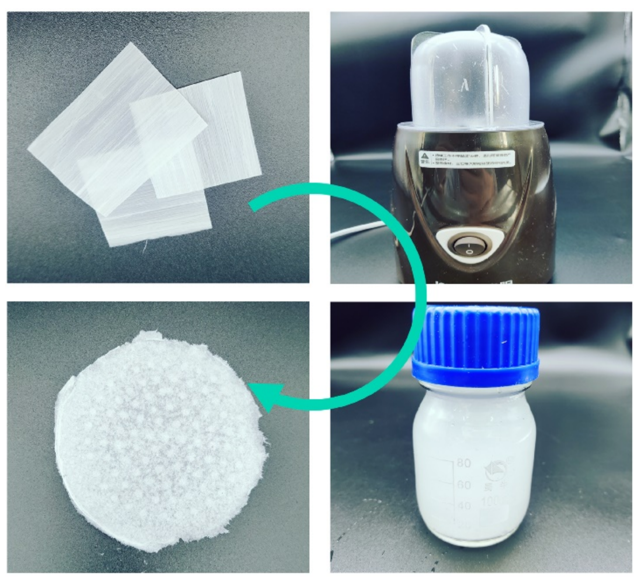

Figure S10. Recycle process by cutting, stirring, and casting.

Table S1. Comparison of density variation and tensile property variation of WSF and other materials prepared by the top-down method.

Resulting material	Raw material	Methods	Density (g cm^{-3})	Increased proportion (times)	Tensile strength (MPa)	Increased proportion (times)	Ref.
WSF	**Wheat straw stem**	**1. Delignification 2. Pressing**	**1.30**	**4-times**	**754**	**18-times**	**This study**
Transparent wood film	**Balsa wood**	**1. Delignification 2. Pressing**	**1.2**	**7-times**	**470**	**26-times**	**5**
Ca^{+} crosslinking wood film	**Balsa wood**	**1. Delignification 2. Tempo-mediated oxidation 3. Ca^{+} crossing linking 4. pressing**	**1.2**	**11-times**	**426**	**31-times**	**6**
Wood film	**Balsa wood**	**1.Partial delignification 2. Pressing**	**-**	**-**	**342**	**20-times**	**7**
Densified wood	**Balsa wood**	**1.Partial delignification 2. Hot-pressing**	**1.3**	**2.8-times**	**587**	**11.5-times**	**8**
High-strength bamboo fiber	**Bamboo**	**1. Delignification 2. Dry**	**-**	**-**	**1900**	**7.2-times**	**9**
Densified bamboo	**Bamboo**	**1. High-pressure steam treatment 2. Partial delignification 3. Pressing**	**1.35**	**1.7-times**	**770**	**2.58-times**	**10**

Table S2. Comparison of tensile properties of WSF and other cellulosed-based materials

Materials	Tensile Strength (Mpa)	Modulus (Gpa)	Toughness (MJ m^{-3})	Ref.
WSF	754.34	22.74	18.72	This work
Densified delignified wood film	350	11.25	7.38	11
Compressed delignified wood	351.8	32.9	4.1	12
Multiscale cellulose fiber film	258	-	7.90	4
Highly aligned Bacterial Cellulose film	1005.3	48.1	24.7	13
CNF/Lignosulfronic acid film	249	4.4	23.6	14
PEG grafted CNF ribbon	254	8.5	15.7	15

REFERENCES

1. M. Frey, L. Schneider, H. Razi, E. Trachsel, E. Faude, S. M. Koch, K. Masania, P. Fratzl, T. Keplinger and I. Burgert, *ACS Sustainable Chemistry & Engineering*, 2021, 9, 9638-9646.
2. X. S. Han, Y. H. Ye, F. Lam, J. W. Pu and F. Jiang, *JOURNAL OF MATERIALS CHEMISTRY A*, 2019, 7, 27023-27031.
3. I. Cave, *Wood science and technology*, 1997, 31, 225-234.
4. Q.-F. Guan, K.-P. Yang, Z.-M. Han, H.-B. Yang, Z.-C. Ling, C.-H. Yin and S.-H. Yu, *ACS Materials Letters*, 2022, 4, 87-92.
5. Q. Fu, Y. Chen and M. Sorieul, *ACS Nano*, 2020, 14, 3528-3538.
6. H. Sun, T. Ji, X. Zhou, H. Bi, M. Xu, Z. Huang and L. Cai, *Materials Chemistry Frontiers*, 2021, 5, 7903-7909.
7. W. Gan, C. Chen, H.-T. Kim, Z. Lin, J. Dai, Z. Dong, Z. Zhou, W. Ping, S. He, S. Xiao, M. Yu and L. Hu, *Nature Communications*, 2019, 10, 5084.
8. J. Song, C. Chen, S. Zhu, M. Zhu, J. Dai, U. Ray, Y. Li, Y. Kuang, Y. Li, N. Quispe, Y. Yao, A. Gong, U. H. Leiste, H. A. Bruck, J. Y. Zhu, A. Vellore, H. Li, M. L. Minus, Z. Jia, A. Martini, T. Li and L. Hu, *Nature*, 2018, 554, 224-228.
9. Z. Li, C. Chen, H. Xie, Y. Yao, X. Zhang, A. Brozena, J. Li, Y. Ding, X. Zhao, M. Hong, H. Qiao, L. M. Smith, X. Pan, R. Briber, S. Q. Shi and L. Hu, *Nature Sustainability*, 2022, 5, 235-244.
10. Z. Li, C. Chen, R. Mi, W. Gan, J. Dai, M. Jiao, H. Xie, Y. Yao, S. Xiao and L. Hu, *Advanced Materials*, 2020, 32, 1906308.
11. M. Zhu, Y. Wang, S. Zhu, L. Xu, C. Jia, J. Dai, J. Song, Y. Yao, Y. Wang and Y. Li, *Advanced Materials*, 2017, 29, 1606284.
12. X. Han, Y. Ye, F. Lam, J. Pu and F. Jiang, *Journal of Materials Chemistry A*, 2019, 7, 27023-27031.
13. S. Wang, T. Li, C. Chen, W. Kong, S. Zhu, J. Dai, A. J. Diaz, E. Hitz, S. D. Solares, T. Li

and L. Hu, 2018, 28, 1707491.
14. J. Zhou, Z. Fang, J. Cui, X. Zhang, Y. Qian, W. Liu, D. Yang and X. Qiu, *Carbohydrate Polymers*, 2021, 259, 117759.
15. H. Tang, N. Butchosa and Q. Zhou, *Advanced materials*, 2015, 27, 2070-2076.