Supplementary material:

Bio-inspired, UV-blocking, water-stable and antioxidant lignin/cellulose films combining high strength, toughness and flexibility

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Sample code	Process parameters			Ultrasonic treatment	Density (g cm^{-1})	$ZnCl_2$ in final films (%)
	Cellulose (g)	Lignin/acetone (mg/ml)	$ZnCl_2/H_2O$ (g/100g)			
C	0.5	0	0	No	1.09	0
UC	0.5	0	0	Yes	1.14	0
UCZn-15	0.5	0	15	Yes	1.14	1.71
UCL	0.5	30	0	Yes	1.17	0
UCLZn-15	0.5	30	15	Yes	1.19	1.63
UCLZn-30	0.5	30	30	Yes	1.19	2.74
UCLZn-45	0.5	30	45	Yes	1.19	3.61

Table S1. Films with different process parameters.

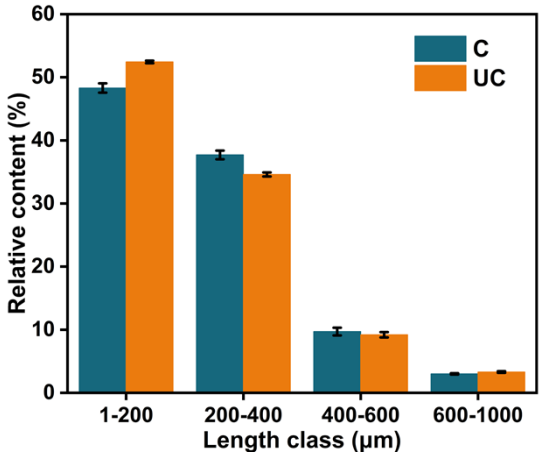

Figure S1. The basic length range of C and UC determined by L&W Fiber tester plus (Sweden).

The MFC length ranges were mainly 1–200 μm (approximately 48.3%) and 200–400 μm (approximately 37.7%). After ultrasonic treatment, the MFC ranges were mainly 1–200 μm (approximately 52.4%) and 200–400 μm (approximately 34.7%).

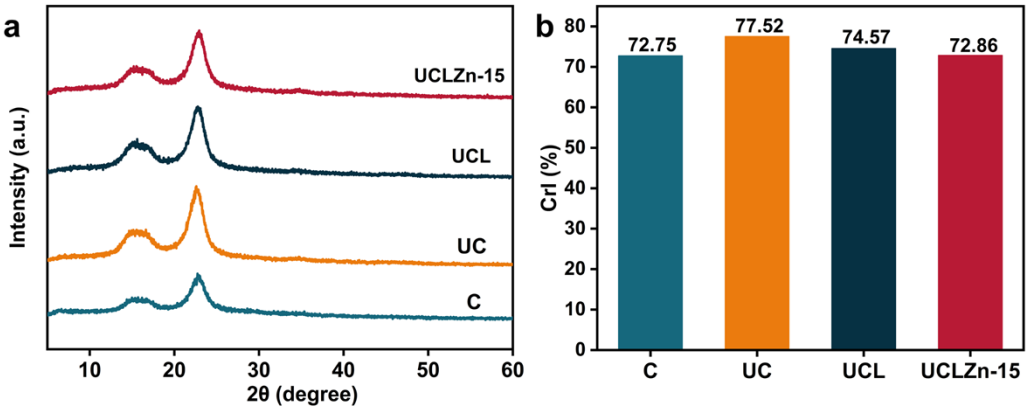

Figure S2 X-ray diffraction pattern (a) and CrI of C, UC, UCL, and UCLZn-15 (b).

To characterize the crystallization of the films, we performed XRD tests. The crystallinity of the cellulose film was 72.75%, whereas the UC films exhibited a high degree of crystallinity of up to 77.52% after ultrasound treatment, which was attributed to the amorphous cellulose removal and degraded of partially amorphous cellulose. After the addition of lignin, the non-crystalline lignin attenuated the crystallinity of the material. With the addition of Zn^{2+}, the crystallinity of the film decreased to 72.86%, illustrating that the crystal structure of the material was changed, which may be attributed to the coordination interactions of cellulose hydroxyl groups and lignin oxygen-containing groups with Zn^{2+}.

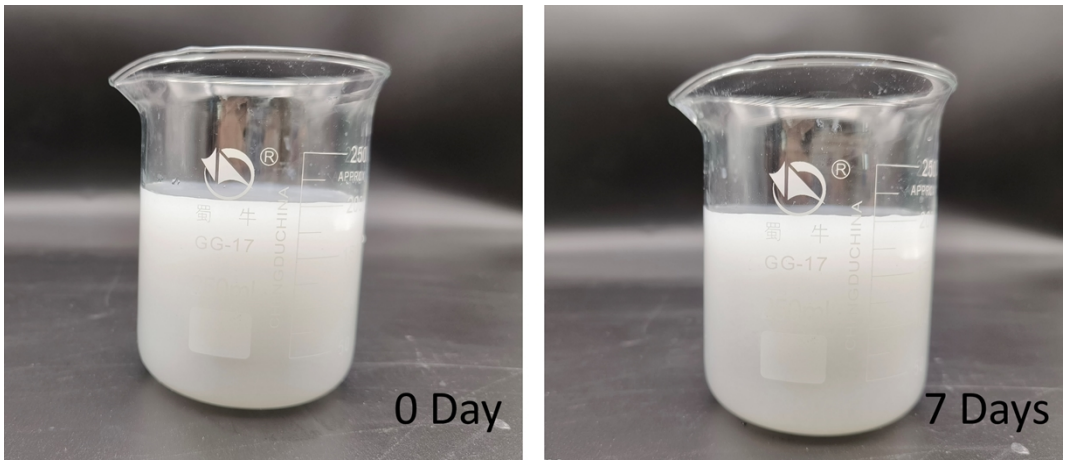

Figure S3. Electronic photo of 0.5%wt ultrasound MFC after sonication.

After sonication, the MFC aqueous suspension has good homogeneity and remains homogeneous without stratification after one week of resting. This is attributed to the higher WRV of the fibers after sonication.

	Strength (Mpa)	Elongation at break (%)	Toughness (MJ/m^3)	Ref
This work	138.14	47.78%	12.88	
lignocellulosic bioplastic	128	-	2.8	1
LM film	10.37	-	11.2	2
cellulose-lignin-glucomannan	110.47	8.27	-	3
cellulose/lignin straw	75.2	-	-	4
lignin-containing cationic wood	70.7	4	-	5
cellulose/lignin Composite Film	74.3-79.2	19	-	6
phosphorylated cellulose nanofibrils	92.7	11.9	7.7	7
cellulose-lignin biodegradable film	92	6	-	8
cellulose/lignin bulk	198	-	8.5	9

Table S2. Films with different process parameters. ("-" Represents not mentioned).

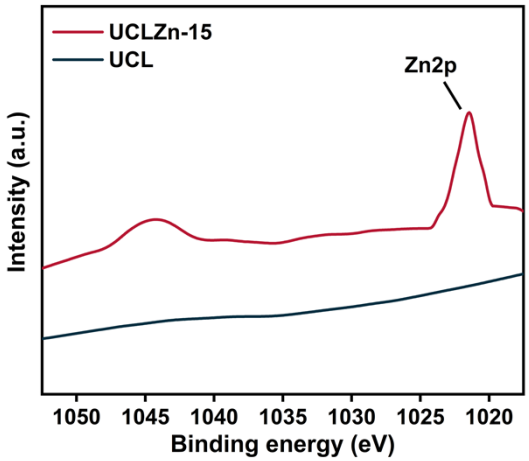

Figure S4 XPS spectra of UCLZn15 and UCL.

Compared to UCL, UCLZn-15 produced a new peak at 1021.7 eV, attributed to the Zn $2p_{2/3}$ binding energy after the addition of Zn^{2+}.

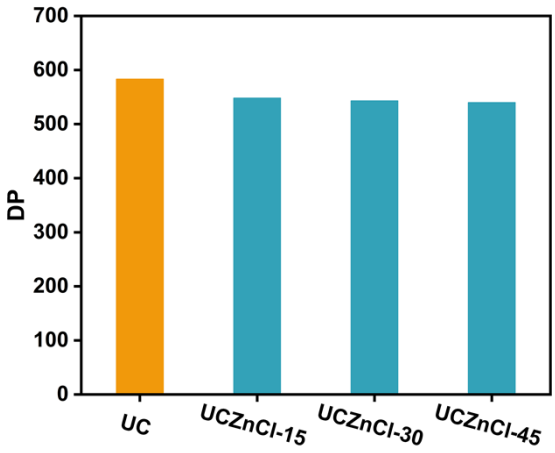

Figure S5 The DP of ultrasonic cellulose before and after $ZnCl_2$ treatment.

The HCl used in the preparation process was mainly used to prevent the hydrolysis of $ZnCl_2$, and the acidic $ZnCl_2$ did not cause serious hydrolysis of cellulose.

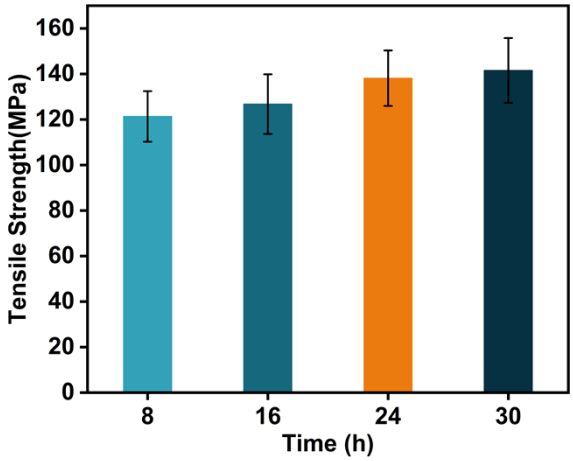

Figure S6 Tensile strength of films with different time of impregnation in the lignin/acetone solution.

The lignin content of the UCZn-15 films gradually increased as the films were impregnated in the lignin/acetone solution for increasing periods of time, reaching 9.78%, 13.2%, 16.8% and 18.1%, respectively. The films impregnated with 24h of lignin/acetone showed the best mechanical properties, indicating that lignin bound best to cellulose at this concentration. Longer impregnation times (>24h) did not significantly improve the lignin content or the mechanical properties.

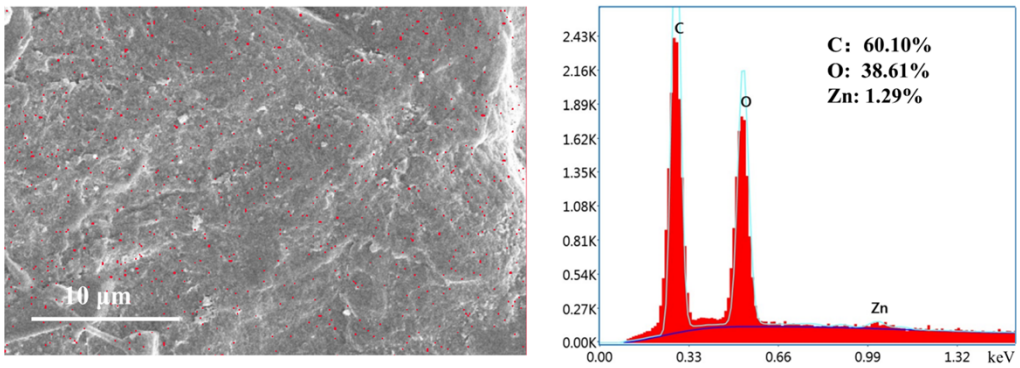

Figure S7 SEM micrographs and corresponding surface layer element scanning results (EDS) of the UCLZn-15.

The characteristic element "Zn" (red spots) was uniformly distributed on the film surface. The relative amounts of C, O, and Zn were 60.10%, 38.61%, and 1.29%, respectively.

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