Supporting Information

High-Density Molded Cellulose Fibers and Transparent Biocomposites Based on Oriented Holocellulose

*Xuan Yang†, Fredrik Berthold‡, and Lars A. Berglund†**

† Wallenberg Wood Science Center, Department of Fiber and Polymer Technology, KTH Royal Institute of Technology, SE-10044 Stockholm, Sweden

‡ RISE, Division of Bioeconomy, Box 5604, Stockholm, Sweden

*Email: blund@kth.se

Table S1. Relative carbohydrate composition of holocellulose fibers and bleached kraft fibers

	Arabinose (%)	Galactose (%)	Glucose (%)	Xylose (%)	Mannose (%)
Fiber-0R	0.62	0.88	77.33	6.66	14.51
Fiber-125R	0.59	0.85	78.35	6.32	13.89
Fiber-250R	0.57	0.84	78.38	6.21	14.01
Fiber-500R	0.56	0.81	78.54	6.10	13.99
K-Fiber-0R	0.55	0.18	84.37	8.26	6.63

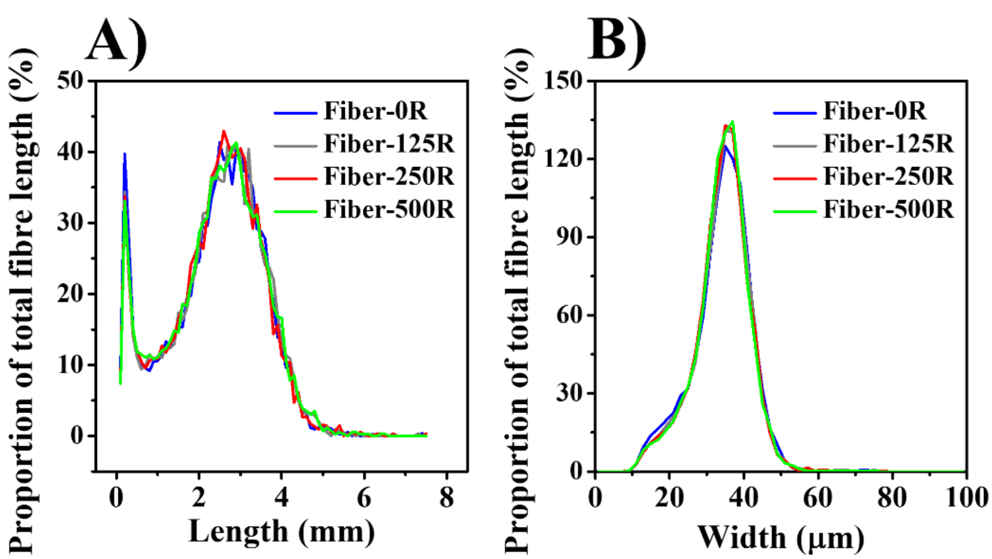

Figure S1. Length (A) and width (B) distributions of holocellulose fibers after different extents of beating.

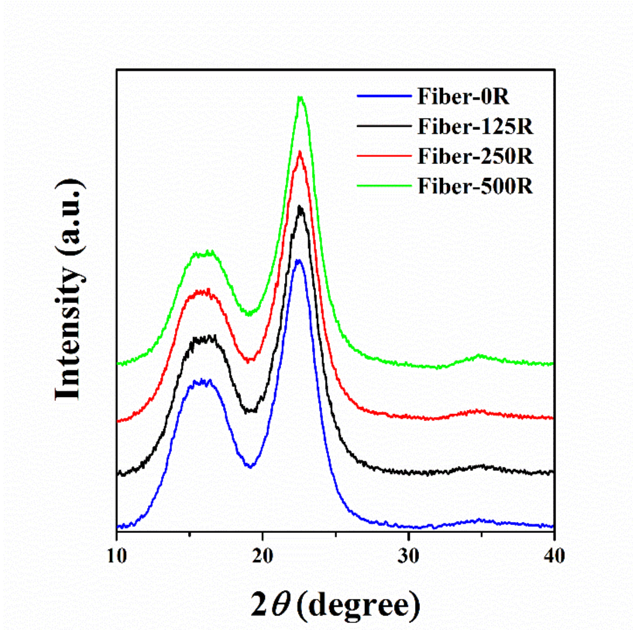

Figure S2. XRD diffractograms of holocellulose fibers after different extents of beating.

Calculation of Fiber Orientation from WAXD results:

Firstly, Hermans orientation parameter was calculated based on the WAXD results, which reflects the orientation of cellulose nanofibrils instead of each single holocellulose fiber. By treating the materials as a laminate structure with a single rotating angle, the Hermans orientation parameter (f) can be defined as

$$f = \frac{3\langle cos^2\varphi\rangle - 1}{2} \qquad \text{- Equation S1}$$

Where φ is the rotating angle of cellulose nanofibrils ($\varphi_{cellulose\ nanofibrils}$). In order to calculate the rotating angle of fibers, the influence of microfibril angle (MFA) needs to be eliminated. In a single fiber, we are treating it as a laminate structure with cellulose nanofibrils as building blocks, while average nanofibrils off-axis angle is actually the MFA. Thus simplified relationship between rotating angle of cellulose nanofibrils ($\varphi_{cellulose\ nanofibrils}$), rotating angle of fibers (φ_{fibers}), and MFA is:

$$\varphi_{fibers} = \varphi_{cellulose\ nanofibrils} - MFA \qquad \text{- Equation S2}$$

The average MFA of holocellulose fibers are estimated to be ~15° according to our previous study on signalized holocellulose fibers (*Yang, X.; Berthold, F.; Berglund, L. A. Preserving Cellulose Structure: Delignified Wood Fibers for Paper Structures of High Strength and Transparency. Biomacromolecules 2018, 19, 3020–3029.*).

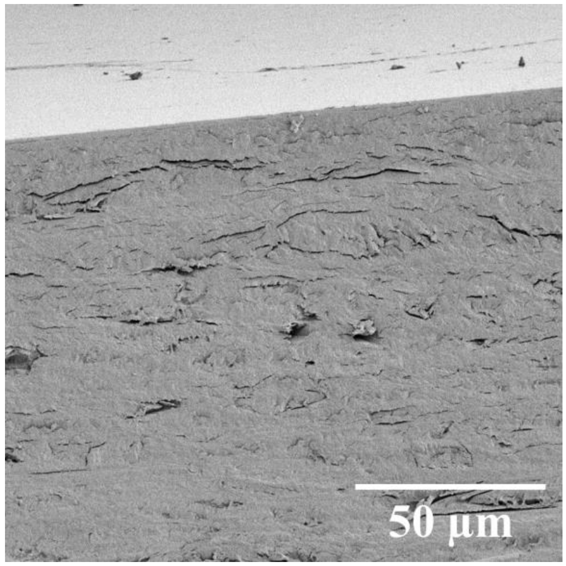

Figure S3. SEM images of freeze-fracture cross-sections of MF-0R/PMMA, showing the good infiltration of MMA throughout the fiber networks.

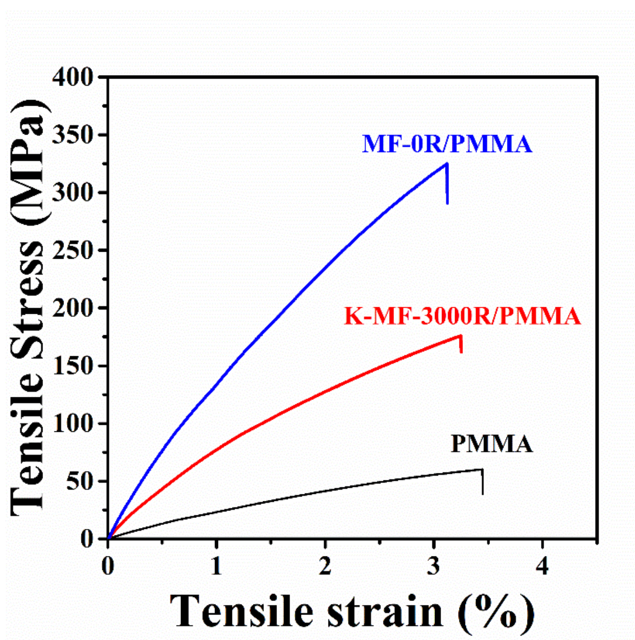

Figure S4. Stress-strain curves of neat PMMA, K-MF-3000R/PMMA biocomposite and MF-0R/PMMA biocomposite at longitudinal direction.