

## Supporting Information

### Synthesis of Modified Lignin as an Antiplasticizer for Strengthening Poly(vinyl alcohol)-Lignin Interactions toward Quality Gel-Spun Fibers

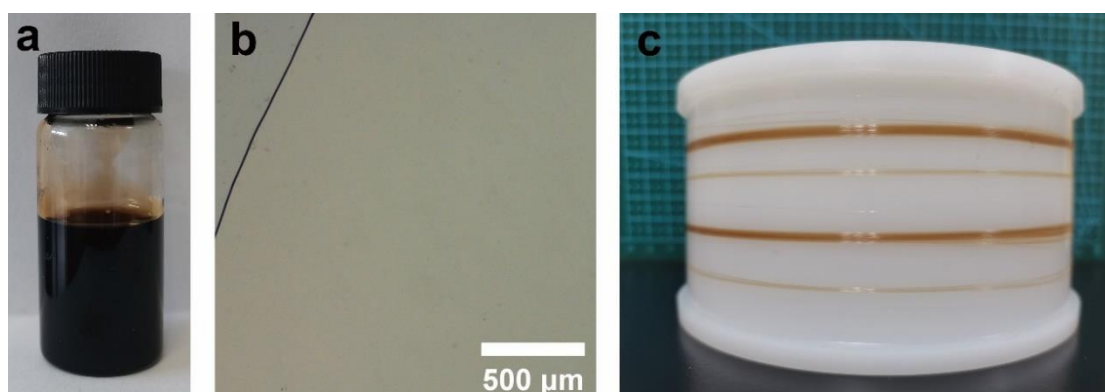
Jiaxian Lin,<sup>1</sup> Yu Cheng,<sup>1</sup> Zheng Li,<sup>2</sup> Yuanyuan Zheng,<sup>1</sup> Bo Xu,<sup>3</sup> and Chunhong Lu<sup>1,\*</sup>

<sup>1</sup> Key Laboratory of Textile Science & Technology, College of Textiles, Donghua University, Shanghai 201620, China

<sup>2</sup> State Key Laboratory for Modification of Chemical Fibers and Polymer Materials, College of Material Science and Engineering, Donghua University, Shanghai 201620, China

<sup>3</sup> College of Chemistry, Chemical Engineering and Biotechnology, Donghua University, Shanghai 201620, China

\*E-mail: chlu@dhu.edu.cn



**Figure S1.** (a) Digital photograph and (b) optical micrograph of 5% ML<sub>24</sub>/PVA spinning dope. (c) The digital photograph of 5% ML<sub>24</sub>/PVA fiber.

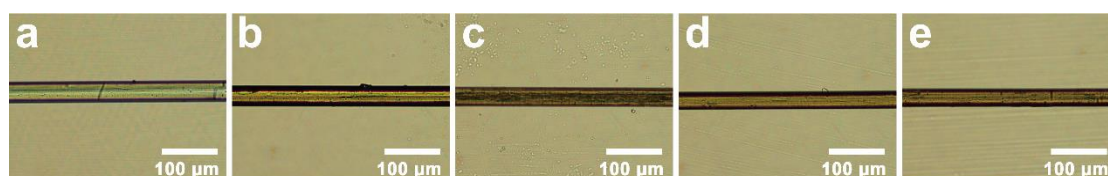
The diameter of fibers was analyzed by optical microscopic images, which were obtained by optical microscopy (ECLIPES LV100N POL, Nikon, Japan). Each fiber type was measured for five times. The density ( $\rho$ ) of each fiber type was calculated by the following equation:

$$\rho = \frac{m}{\pi \times (D/2)^2 \times L} \quad (\text{S1})$$

where  $m$  refers to the mass (g) of fiber with certain length of  $L$  (cm).  $D$  refers to the diameter ( $\mu\text{m}$ ) of the fiber.

**Table S1.** Estimation of the Density of All Gel-Spun Fibers

| Fiber Type                     | Measured Diameter ( $\mu\text{m}$ ) | Calculated Fiber Density ( $\text{g}/\text{cm}^3$ ) |
|--------------------------------|-------------------------------------|---|
| PVA fiber                      | $39.3 \pm 0.8$                      | $1.35 \pm 0.05$                                     |
| 5% OL/PVA fiber                | $37.3 \pm 0.1$                      | $1.32 \pm 0.01$                                     |
| 5% ML <sub>12</sub> /PVA fiber | $35 \pm 0.6$                        | $1.34 \pm 0.04$                                     |
| 5% ML <sub>24</sub> /PVA fiber | $32 \pm 0.4$                        | $1.30 \pm 0.03$                                     |
| 5% ML <sub>48</sub> /PVA fiber | $31.4 \pm 0.6$                      | $1.34 \pm 0.05$                                     |



**Figure S2.** Optical microscopic graphs of (a) neat PVA, (b) 5% OL/PVA, (c) 5% ML<sub>12</sub>/PVA, (d) 5% ML<sub>24</sub>/PVA and (e) 5% ML<sub>48</sub>/PVA fibers.

The conversion ratio of D-gluconic acid was calculated by the following equation:

$$\text{Conversion ratio (\%)} = \frac{W_1 - W_0}{W_2} \times 100\% \quad (\text{S2})$$

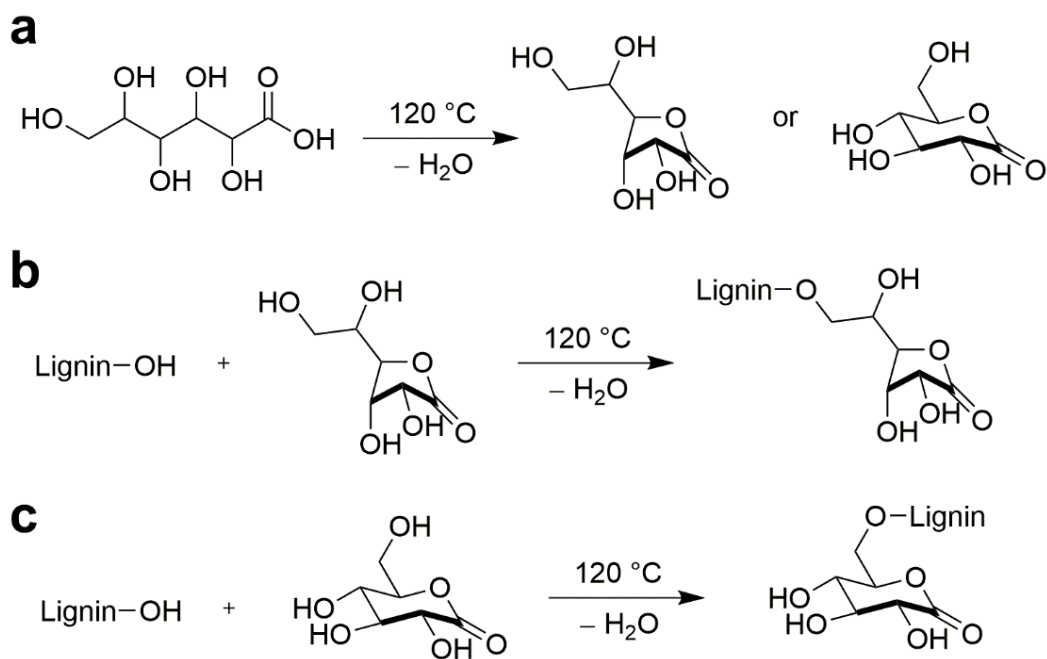
where  $W_1$  is the actual weight of modified lignin,  $W_0$  is the initial weight of unmodified lignin,  $W_2$  is the actual weight of D-gluconic acid in the 20 mL of D-gluconic acid aqueous solution.

**Table S2.** The Chemical Reaction Conditions and Conversion Ratios of All Lignin

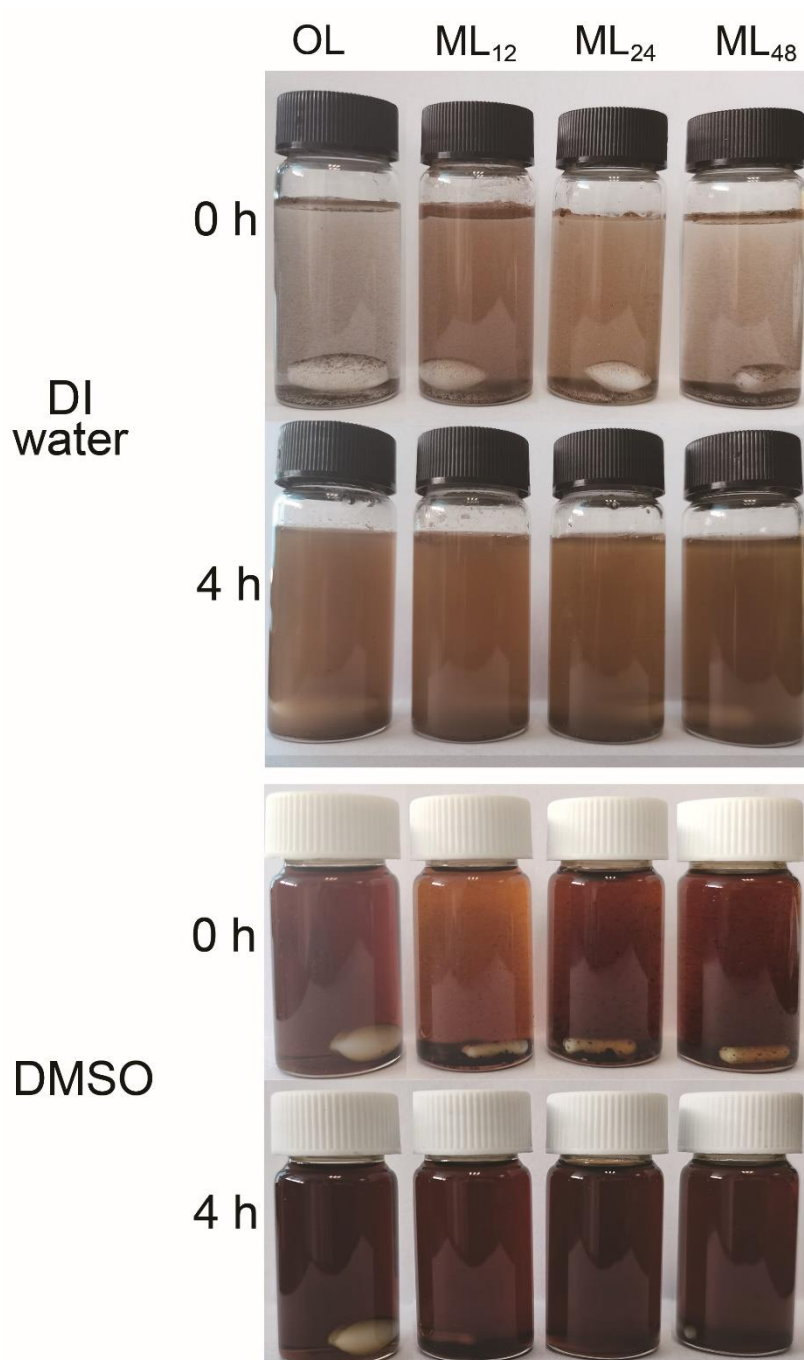
Samples

| Lignin Type      | Reaction Time (h) | Conversion Ratio (%) |
|------------------|-------------------|----------------------|
| OL               | N/A               | N/A                  |
| ML <sub>12</sub> | 12                | 3.9                  |
| ML <sub>24</sub> | 24                | 10.2                 |
| ML <sub>48</sub> | 48                | 15.7                 |

N/A: not applicable



**Figure S2.** Possible side reactions during lignin modification, including (a) lactonization of D-gluconic acid to yield D-glucono-1,4-lactone and D-glucono-1,5-lactone, etherification of lignin with (b) D-glucono-1,4-lactone and (c) D-glucono-1,5-lactone, respectively.



**Figure S3.** Solubility measurements of OL, ML<sub>12</sub>, ML<sub>24</sub> and ML<sub>48</sub> in DI water (concentration of 1 mg mL<sup>-1</sup>) and DMSO (concentration of 2 mg mL<sup>-1</sup>), respectively.

**Table S2.** Mechanical Properties of 5% OL/PVA Fibers from 100% DMSO and DMSO/H<sub>2</sub>O (80/20, v/v) Solvents

| Fiber Sample   | Draw Ratio | Tensile Strength (MPa) | Young's Modulus (GPa) | Toughness (J/g) |
|--|------------|------------------------|-----------------------|-----------------|
| 5% OL/PVA fibers from 100% DMSO                          | 30.1       | $890 \pm 16$           | $14.7 \pm 1.3$        | $20.8 \pm 0.8$  |
| 5% OL/PVA fibers from DMSO/H <sub>2</sub> O (80/20, v/v) | 23.9       | $697 \pm 67$           | $15.0 \pm 1.6$        | $13.2 \pm 2.1$  |