

## **High Lignin-containing Nanocelluloses Prepared via TEMPO-mediated Oxidation and Polyethylenimine Functionalization for Antioxidant and Antibacterial Applications**

Yisheng Sun <sup>a</sup>, Hanwen Zhang <sup>a</sup>, Qianwei Li <sup>a</sup>, Bongkosh Vardhanabhuti <sup>b</sup>, Caixia Wan <sup>a,\*</sup>

<sup>a</sup> Department of Biomedical, Biological, and Chemical Engineering, University of Missouri,  
Columbia, Missouri 65211, USA

<sup>b</sup> Division of Food, Nutrition & Exercise Sciences, University of Missouri, Columbia, Missouri  
65211, USA

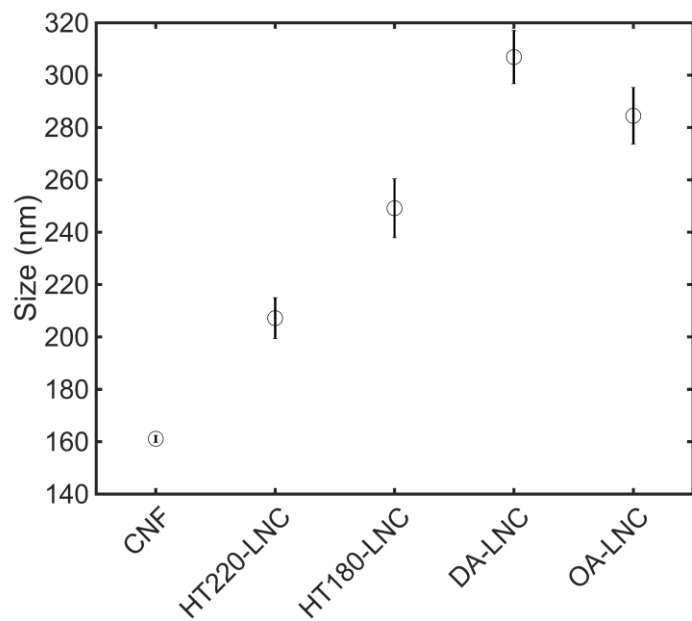
\* Corresponding author: Phone: +1 573 884 7882; E-mail: [wanca@missouri.edu](mailto:wanca@missouri.edu)

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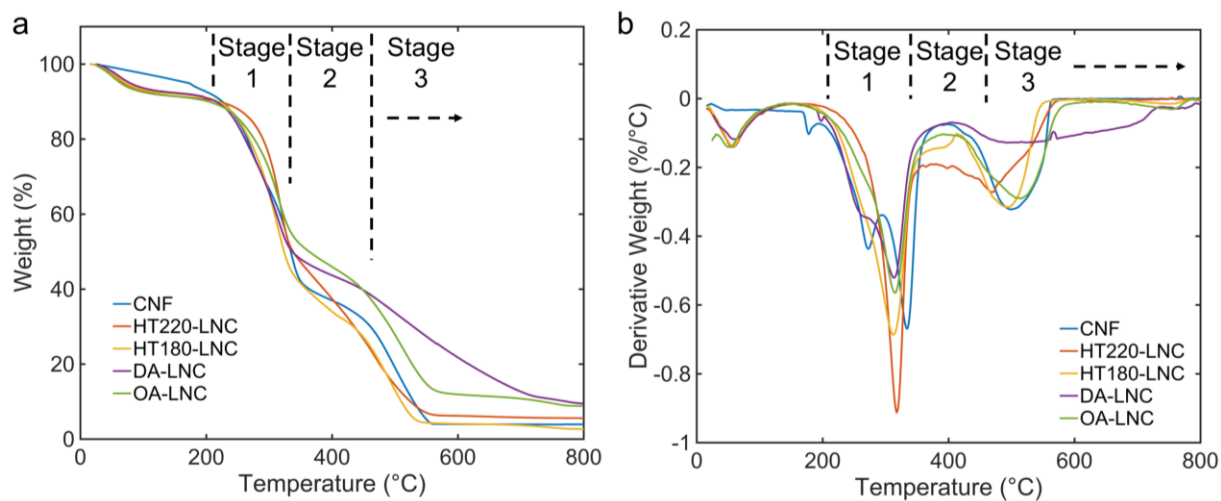
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## Experimental method

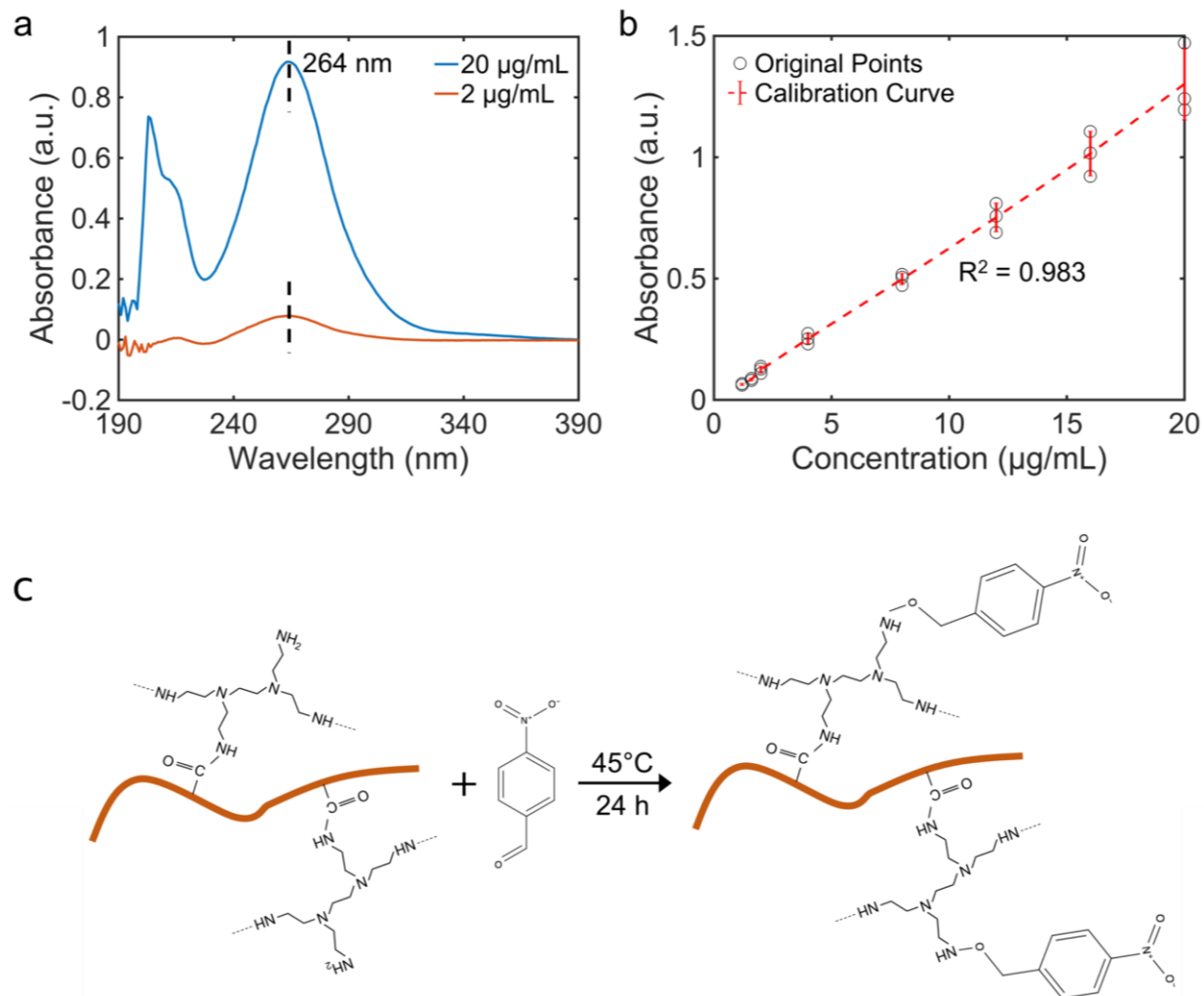
*Quantification of primary amino (-NH<sub>2</sub>) groups.* PEI-modified nanocellulose suspensions were centrifuged at 5000 ×g for 5 min to remove water and then washed by pure methanol via the same centrifugation for 4 times. In the end, the PEI-modified nanocelluloses were resuspended in pure methanol with consistency of 2 mg/mL. Meanwhile, 4-nitrobenzaldehyde was dissolved in pure methanol at 20 mg/mL as a stock solution. Gradient dilution was then applied and the characteristic absorbance at 264 nm was measured by a Cary 50 spectrophotometer (Varian, CA, US) to create a linear calibration curve (Fig. S3b). The reaction between PEI-modified nanocelluloses and 4-nitrobenzaldehyde was carried out by mixing 2.5 mL of nanocellulose suspension and 10 mL of 100 μg/mL 4-nitrobenzaldehyde solution followed by the reaction at 45 °C with 500 rpm shaking for 24 h. After reaction, the mixtures were filtrated by 0.45 μm syringe filter and their absorbances were measured at 264 nm to determine the consumption of 4-nitrobenzaldehyde by PEI-modified nanocelluloses compared to the control using pure methanol. The primary amine content in PEI-modified nanocelluloses was determined by reacting 4-nitrobenzaldehyde with primary amine groups by a 1:1 molar ratio (Fig. S3c).



**Fig. S1.** Nanocellulose sizes by dynamic light scattering (DLS) analysis.



**Fig. S2.** TGA analysis of nanocelluloses. (a) Mass change over temperature. (b) Derivative mass change over temperature.



**Fig. S3.** Determination of primary amino groups. (a) UV-vis spectra of 4-nitrobenzaldehyde. (b) Calibration curve of the absorbance of 4-nitrobenzaldehyde at 264 nm. (c) Schematic reaction mechanism between PEI-modified nanocelluloses and 4-nitrobenzaldehyde.