Supplemental information

A Sequential Autohydrolysis-Ionic Liquid Fractionation Process for High Quality Lignin Production

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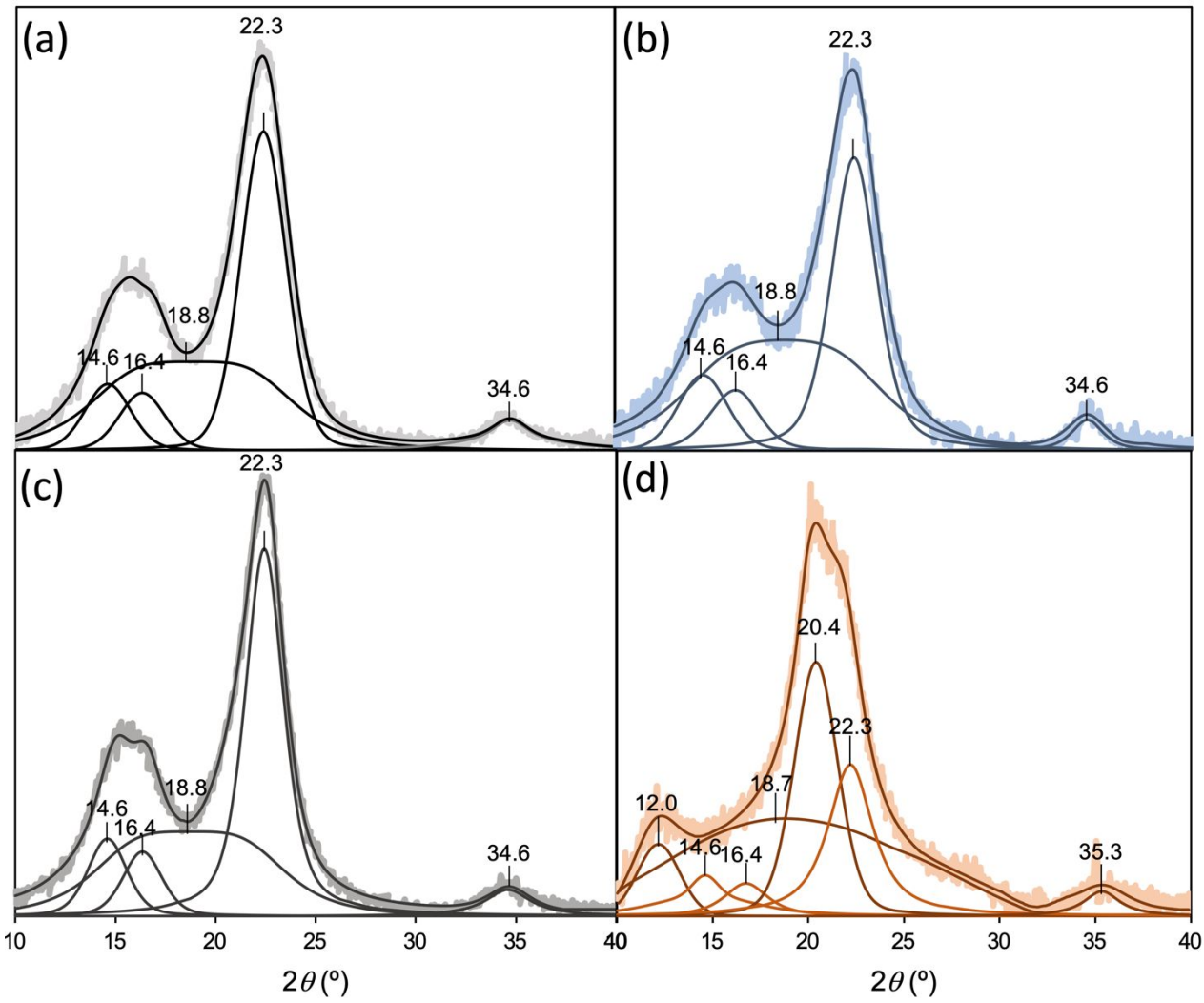

Figure S1. Deconvoluted X-ray diffractograms of hybrid poplar wood powder using Pseudo-Voigt function. The amorphous peaks were fitted using InvsPoly function in OriginPro 2020. All peaks were fitted until they converged at $R^2 = 0.99$. Legend: (a) Untreated hybrid poplar (HP); (b) IL-activated HP; (c) Autohydrolyzed HP; (d) Autohydrolyzed + IL-activated HP. Autohydrolysis pretreatment was performed at 160 °C for 60 min and IL-activation was performed with 1-ethyl-3-methylimidazolium acetate at 60 °C for 3 h.

Table S1. Calculation of Cellulose Crystallinity Index (CrI) of Pretreated and Untreared Hybrid Poplar Wood Powder following the Peak Deconvolution Method.

Sample name	**Total Cellulose-I crystalline area***	**Total Cellulose-II crystalline area***	**Amorphous area***	**% Cellulose-I crystallinity†**	**% Cellulose-II crystallinity†**
Control	1089	n/a	798	58	n/a
IL-activated	797	n/a	726	52	n/a
Autohydrolyzed	1408	n/a	975	59	n/a
Autohydrolyzed+IL-activated	458	572	798	25	31

†Crystallinity index (%) was calculated using the formula $\frac{Peak\ area_{crystalline\ cellulose\ I\ or\ II}}{Area_{crystalline\ cellulose\ I} + Area_{crystalline\ cellulose\ II} + Area_{amorphous}} \times 100$

*Peak areas were determined after baseline treatment and integration of delimited area using the Peak Analyzer dialog box in OriginPro 2020 software.

Table S2. Semi-quantitative Estimation of Inter-unit Linkages in Hybrid Poplar Lignin using 2D HSQC NMR Analysis.

Inter-unit linkage	**Chemical shift δC/δH (ppm)**	**Native biomass**†	**Ionic liquid (IL)-lignin**†
Aryl ether (β-O-4)	72.7/5.0	27.3	24.4
Phenylcoumaran (β-5)	87.4/5.7	3.1	1.1
Resinol (β-β')	85.7/4.7	4.6	6.1
Total ether side chains	n/a	35.0	31.7
p-Hydroxybenzoate (P)	131.7/7.8	4.9	3.9
Stilbene (SB)	127.9/7.0	0	1.9

† The number of inter-unit linkages are expressed as, $\frac{\int X}{\left(\frac{\int S_{2,6}}{2}\right) + \int G_2} \times 100$; where P and SB integral values are obtained by dividing the cross-peak intensities by 2.