Supporting information

for

Cationic Lignin as An Efficient and Sustainable Homogenous Catalyst for Aqueous Knoevenagel Condensation Reactions

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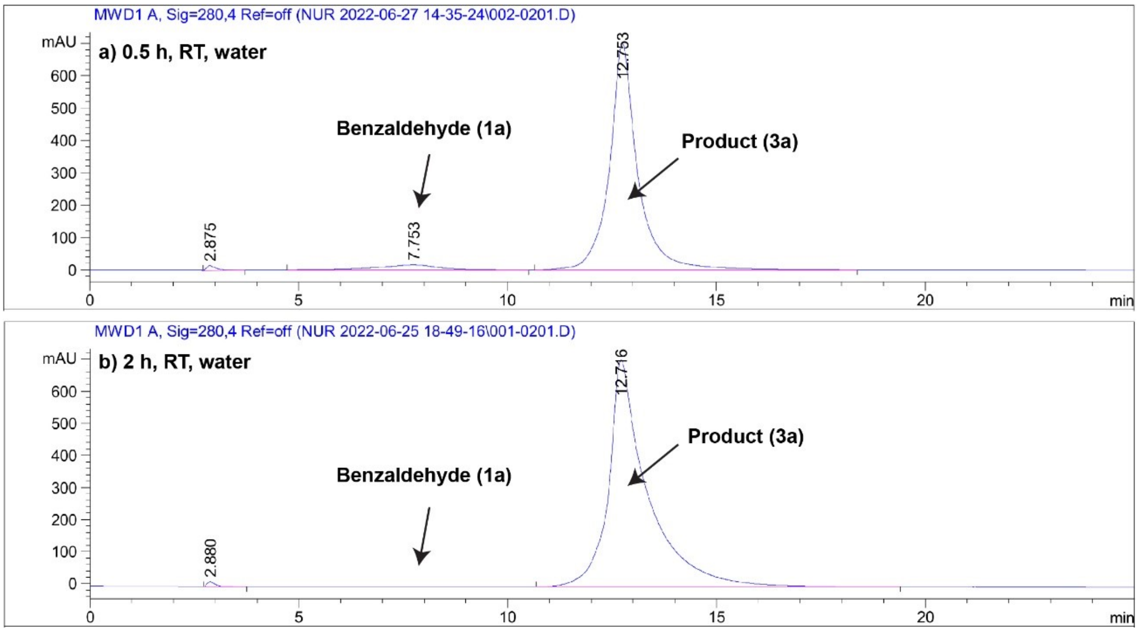

Figure S1. HPLC chromatographs of the Knoevenagel reaction in water at RT and at a) 1h and b) 2h

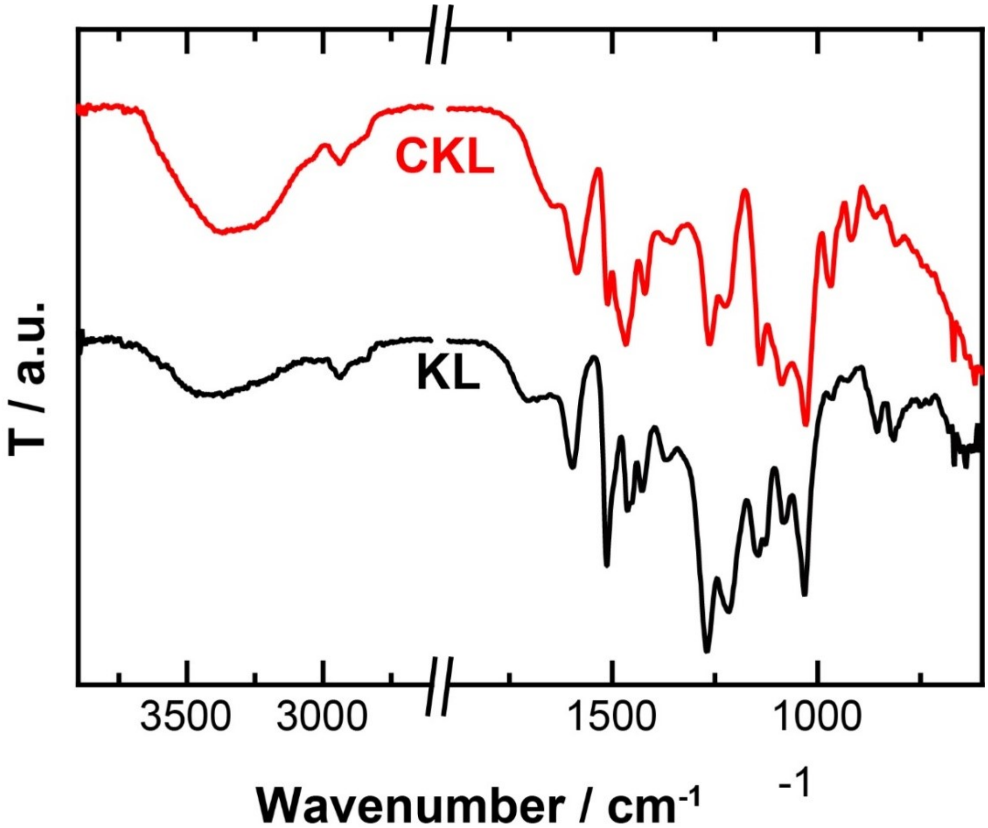

Figure S2. The FTIR spectra of KL and CKL.

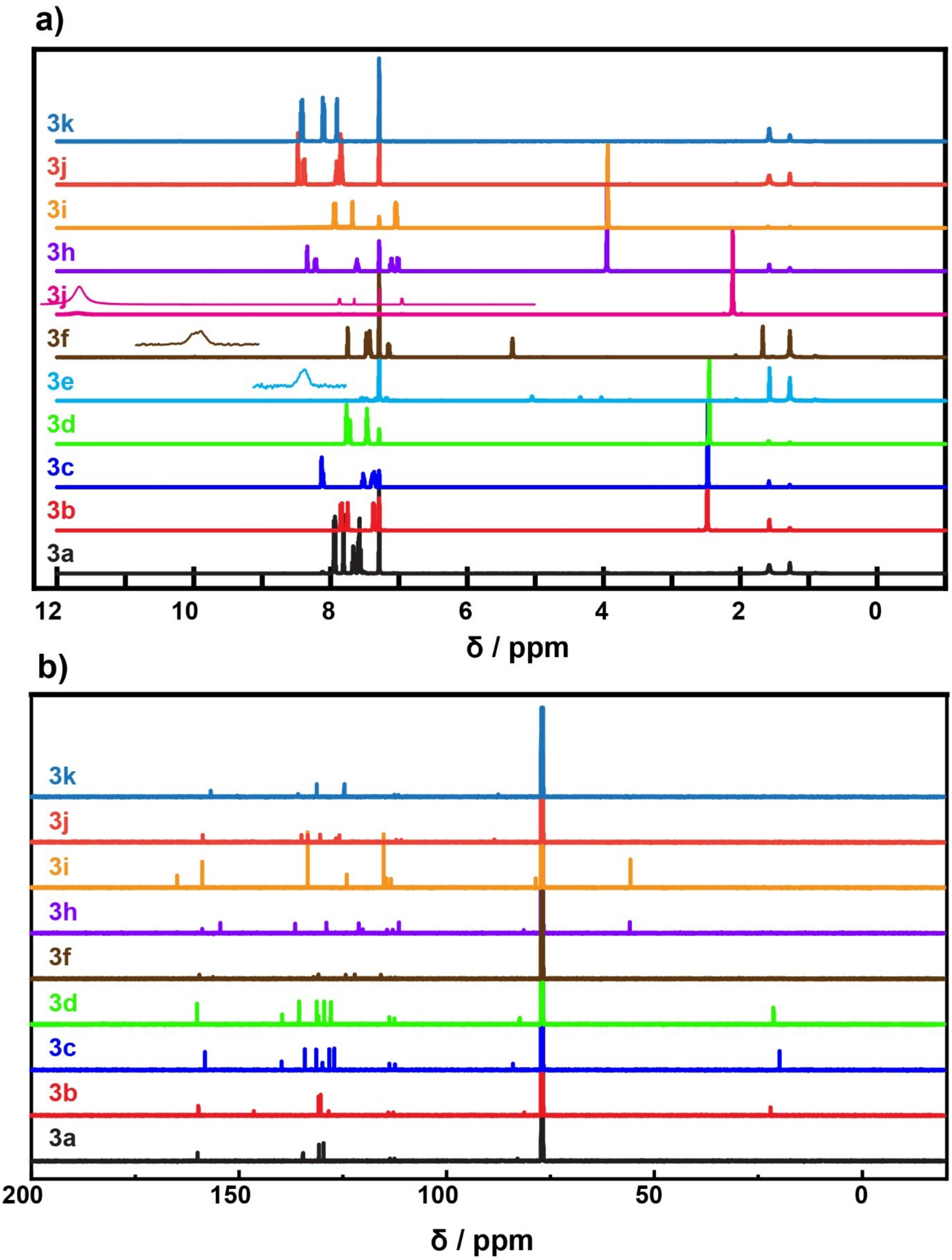

Figure S3. The a) ^{1}H and b) ^{13}CNMR spectra of benzylidenemalononitriles.

<u>Analytical data of the products</u>

- **Compound (3a)**. 1H NMR (500 MHz, CDCl3) δ 1H NMR (500 MHz, Chloroform-d) δ 7.93 (d, 2H), 7.81 (s, 1H), 7.71 – 7.62 (m, 2H), 7.57 (m, 1H). 13C NMR 13C NMR δ 159.95, 134.67, 130.95, 130.76, 129.67, 113.72, 112.55, 82.93.
- **Compound (3b)**. ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.78 – 7.73 (m, 2H), 7.71 (s, 1H), 7.46 (t, J = 3.9 Hz, 2H), 2.46 (d, J = 2.3 Hz, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 160.17, 139.67, 135.60, 131.30, 130.97, 129.54, 127.95, 113.86, 112.66, 82.49, 21.31.
- **Compound (3c)**. ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.15 – 8.09 (m, 2H), 7.52 (t, J = 7.7 Hz, 1H), 7.42 – 7.33 (m, 2H), 2.48 (d, J = 2.4 Hz, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 158.17, 139.75, 134.21, 131.43, 129.92, 128.33, 127.10, 113.84, 112.47, 84.01, 19.86.
- **Compound (3d)**. ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.87 – 7.81 (m, 2H), 7.75 (s, 1H), 7.36 (d, 2H), 2.49 (d, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 159.78, 146.41, 130.95, 130.41, 128.48, 114.04, 112.88, 81.27, 22.06.
- **Compound (3e)**. ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.29 (s, 1H), 7.54 (d, 1H), 7.45 (t, 1H), 7.33 (t, 1H), 7.18 (d, 1H), Some adsorbed CKL might cause other signals.
- **Compound (3f)**. ^{1}H NMR (500 MHz, Chloroform-*d*) δ 9.99 (s, OH), 7.75 (s, 1H), 7.52 – 7.40 (m, 3H), 7.14 (d, 1H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 159.63, 156.30, 132.16, 130.97, 124.30, 122.18, 115.94, 113.61. Some adsorbed CKL might cause other signals.
- **Compound (3g)**. ^{1}H NMR (500 MHz, Chloroform-*d*) δ 11.70 (s, OH), 7.95 – 7.76 (m, 2H), 7.66 (d, 1H), 7.07 – 6.87 (m, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 178.14, 162.27, 159.05, 133.79, 123.79, 116.73, 114.47, 113.39, 78.12, 20.85. Some adsorbed CKL might cause other signals

- **Compound (3h)**. ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.34 (s, 1H), 8.22 (d, 1H), 7.61 (t, 1H), 7.11 (t, 1H), 7.01 (d, 1H), 3.95 (d, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 158.95, 154.53, 136.52, 128.93, 121.26, 120.23, 114.37, 113.02, 111.48, 81.48, 55.96.
- **Compound (3i)**. ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.94 (d, 2H), 7.68 (s, 1H), 7.04 (d, 2H), 3.94 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 164.84, 158.90, 133.50, 124.04, 115.16, 114.47, 113.38, 78.59, 55.84.
- **Compound (3j).** ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.48 (s, 1H), 8.39 (d, 1H), 7.94 – 7.87 (m, 3H). ^{13}C NMR. δ 158.72, 134.96, 133.43, 130.50, 126.72, 125.90, 112.20, 110.97, 88.63.
- **Compound (3k)**. ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.42 (d, 2H), 8.10 (d, 2H), 7.91 (s, 1H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 156.85, 135.79, 131.34, 124.68, 112.63, 111.61, 87.59.