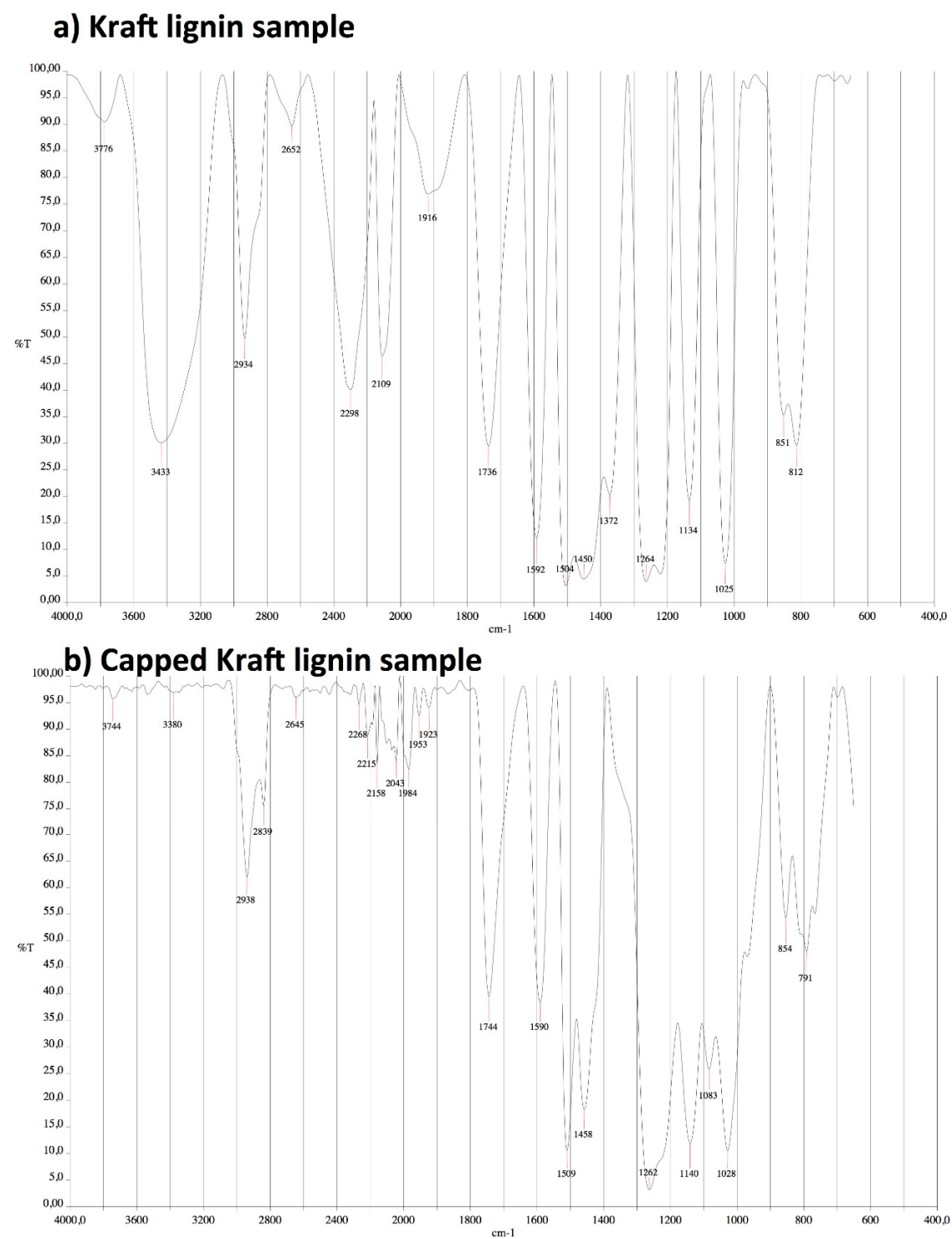


Supplementary Materials: Mechanochemical Lignin-Mediated Strecker Reaction

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1. Figure S1**Figure S1.** IR spectra of Kraft lignin and capped Kraft lignin.

2. Figure S2

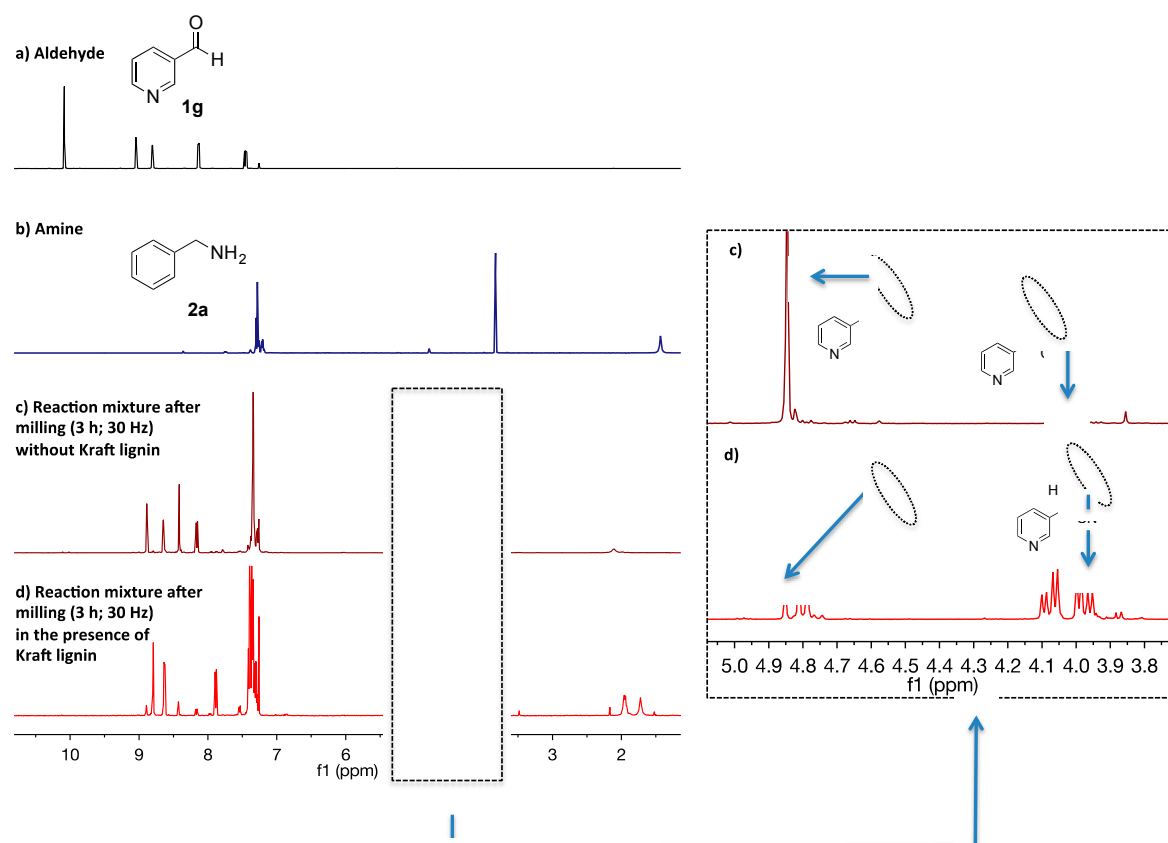


Figure S2. Monitoring of the mechanochemical Strecker reaction of **1g**, **2a**, and KCN by ^1H NMR spectroscopy.

3. Characterization of the Products **3a-j**, **5a** and **6a**2-(Benzylamino)-2-phenylacetonitrile (**3a**) [1,2]

^1H NMR (600 MHz, CDCl_3) δ (ppm) = 7.56 (dd, $J = 7.9$ Hz, $J = 1.4$ Hz, 2H), 7.45–7.40 (m, 4H), 7.40–7.36 (m, 3H), 7.33–7.29 (m, 1H), 4.76 (s, 1H), 4.08 (d, $J = 13.0$ Hz, 1H), 3.97 (d, $J = 13.0$ Hz, 1H), 1.89 (br s, 1H).

^{13}C NMR (150 MHz, CDCl_3) δ (ppm) = 138.2, 134.8, 129.1, 129.0, 128.7, 128.5, 127.7, 127.4, 118.8, 53.5, 51.3.

2-(Benzylamino)-2-(*p*-tolyl)acetonitrile (**3b**) [1,3]

^1H NMR (600 MHz, CDCl_3) δ (ppm) = 7.42 (dd, $J = 7.5$ Hz, $J = 5.7$ Hz, 4H), 7.37 (ddd, $J = 7.6$ Hz, $J = 6.6$ Hz, $J = 1.4$ Hz, 2H), 7.31 (ddd, $J = 6.7$ Hz, $J = 6.3$ Hz, $J = 1.6$ Hz, 1H), 7.22 (d, $J = 7.9$ Hz, 2H), 4.72 (s, 1H), 4.06 (d, $J = 13.0$ Hz, 1H), 3.95 (d, $J = 13.1$ Hz, 1H), 2.38 (s, 3H), 1.87 (br. s, 1H).

^{13}C NMR (150 MHz, CDCl_3) δ (ppm) = 139.1, 138.3, 131.9, 129.7, 128.8, 128.5, 127.7, 127.3, 119.0, 53.3, 51.4, 21.3.

2-(Benzylamino)-2-(2-methoxyphenyl)acetonitrile (**3c**) [4]

^1H NMR (600 MHz, CDCl_3) δ (ppm) = 7.41 (d, $J = 8.0$ Hz, 2H), 7.39–7.35 (m, 4H), 7.33–7.29 (m, 1H), 7.00 (td, $J = 7.5$ Hz, $J = 1.0$ Hz, 1H), 6.95 (dd, $J = 8.1$ Hz, $J = 1.0$ Hz, 1H), 4.83 (s, 1H), 4.09 (d, $J = 13.0$ Hz, 1H), 3.95 (d, $J = 13.0$ Hz, 1H), 3.88 (s, 3H), 2.30 (br s, 1H).

^{13}C NMR (150 MHz, CDCl_3) δ (ppm) = 156.9, 138.4, 130.6, 128.9, 128.6, 128.5, 127.6, 123.3, 121.0, 119.1, 111.3, 55.7, 51.6, 49.4.

2-(Benzylamino)-2-(4-fluorophenyl)acetonitrile (3d) [5]

¹H NMR (600 MHz, CDCl₃) δ (ppm) = 7.57–7.51 (m, 2H), 7.43–7.39 (m, 2H), 7.39–7.34 (m, 2H), 7.33–7.28 (m, 1H), 7.13–7.07 (m, 2H), 4.74 (s, 1H), 4.06 (d, *J* = 12.9 Hz, 1H), 3.95 (d, *J* = 12.9 Hz, 1H), 1.89 (br s, 1H).

¹³C NMR (150 MHz, CDCl₃) δ (ppm) = 163.1 (d, *C*_{ipso}-F, ¹*J*_{CF} = 248.2 Hz), 138.0, 130.7 (d, *C*_{para}-F, ⁴*J*_{CF} = 3.1 Hz), 129.3 (d, *C*_{meta}-F, ³*J*_{CF} = 8.5 Hz), 128.8, 128.5, 127.8, 118.7, 116.0 (d, *C*_{ortho}-F, ²*J*_{CF} = 21.6 Hz), 52.8, 51.3.

2-(Benzylamino)-2-(4-chlorophenyl)acetonitrile (3e) [6]

¹H NMR (400 MHz, CDCl₃) δ (ppm) = 7.54–7.46 (m, 2H), 7.43–7.39 (m, 2H), 7.39–7.25 (m, 5H), 4.73 (s, 1H), 4.05 (d, *J* = 13.0 Hz, 1H), 3.95 (d, *J* = 13.0 Hz, 1H), 1.89 (br s, 1H).

¹³C NMR (100 MHz, CDCl₃) δ (ppm) = 138.0, 135.2, 133.3, 129.3, 128.8, 128.8, 128.5, 127.9, 118.5, 52.9, 51.3.

2-(Benzylamino)-2-(4-bromophenyl)acetonitrile (3f) [6]

¹H NMR (400 MHz, CDCl₃) δ (ppm) = 7.58–7.52 (m, 2H), 7.47–7.33 (m, 6H), 7.33–7.27 (m, 1H), 4.72 (s, 1H), 4.05 (d, *J* = 12.9 Hz, 1H), 3.95 (d, *J* = 13.0 Hz, 1H), 1.89 (br s, 1H).

¹³C NMR (100 MHz, CDCl₃) δ (ppm) = 138.0, 133.9, 132.2, 129.1, 128.8, 128.5, 127.9, 123.3, 118.4, 53.0, 51.3.

2-(Benzylamino)-2-(pyridin-3-yl)acetonitrile (3g) [7,8]

¹H NMR (600 MHz, CDCl₃) δ (ppm) = 8.77 (d, *J* = 2.4 Hz, 1H), 8.60 (dd, *J* = 4.8 Hz, 1.7 Hz, 1H), 7.87 (ddd, *J* = 8.0 Hz, *J* = 2.9 Hz, *J* = 1.4 Hz, 1H), 7.41–7.37 (m, 2H), 7.37–7.32 (m, 3H), 7.31–7.27 (m, 1H), 4.78 (s, 1H), 4.05 (d, *J* = 13.0 Hz, 1H), 3.95 (d, *J* = 12.9 Hz, 1H), 2.06 (br s, 1H).

¹³C NMR (150 MHz, CDCl₃) δ (ppm) = 150.4, 148.8, 137.7, 135.0, 130.7, 128.8, 128.5, 127.9, 123.7, 117.8, 51.3, 51.3.

2-(Benzylamino)-2-cyclohexylacetonitrile (3h) [8]

¹H NMR (400 MHz, CDCl₃) δ (ppm) = 7.43–7.26 (m, 5H), 4.08 (d, *J* = 13.0 Hz, 1H), 3.82 (d, *J* = 13.0 Hz, 1H), 3.31 (d, *J* = 6.3 Hz, 1H), 1.94–1.62 (m, 6H), 1.34–1.07 (m, 5H).

¹³C NMR (100 MHz, CDCl₃) δ (ppm) = 138.6, 128.6, 128.4, 127.6, 119.7, 55.6, 51.9, 40.8, 29.8, 29.0, 26.1, 25.8, 25.7.

2-Phenyl-2-(phenylamino)acetonitrile (3i) [2,6]

¹H NMR (400 MHz, CDCl₃) δ (ppm) = 7.65–7.58 (m, 2H), 7.51–7.41 (m, 3H), 7.32–7.25 (m, 2H), 6.95–6.89 (m, 1H), 6.82–6.76 (m, 2H), 5.44 (s, 1H), 4.05 (br s, 1H).

¹³C NMR (100 MHz, CDCl₃) δ (ppm) = 144.8, 134.1, 129.7, 129.7, 129.5, 127.4, 120.4, 118.3, 114.3, 50.4.

1-(Benzylamino)cyclohexane-1-carbonitrile (3j) [9]

¹H NMR (400 MHz, CDCl₃) δ (ppm) = 7.42–7.25 (m, 5H), 3.92 (s, 2H), 2.11–2.01 (m, 2H), 1.85–1.76 (m, 2H), 1.73–1.51 (m, 5H), 1.37–1.22 (m, 1H).

¹³C NMR (100 MHz, CDCl₃) δ (ppm) = 139.5, 128.7, 128.5, 127.5, 122.3, 57.5, 48.7, 36.2, 25.3, 22.4.

N-Benzylphthalimide (5a) [10]

¹H NMR (400 MHz, CDCl₃) δ (ppm) = 7.85 (dd, *J* = 5.4, 3.1 Hz, 2H), 7.70 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.46–7.41 (m, 2H), 7.37–7.27 (m, 3H), 4.85 (s, 2H).

¹³C NMR (100 MHz, CDCl₃) δ (ppm) = 168.2, 136.5, 134.1, 132.3, 128.8, 128.7, 127.9, 123.4, 41.7.

MS (EI, 70 eV) *m/z* (%) = 237.9 (10) [M⁺], 236.9 (50), 104.0 (100), 91 (27), 77.1 (62).

2-Benzyl-3-oxoisindoline-1-carbonitrile (6a) [11]

¹H NMR (400 MHz, CDCl₃) δ (ppm) = 7.84 (d, *J* = 6.7 Hz, 1H), 7.59–7.46 (m, 3H), 7.31–7.14 (m, 5H), 5.40 (d, *J* = 15.1 Hz, 1H), 4.98 (s, 1H), 4.20 (d, *J* = 15.0 Hz, 1H).

¹³C NMR (100 MHz, CDCl₃) δ (ppm) = 167.0, 136.9, 135.3, 133.1, 131.2, 130.5, 129.3, 128.7, 128.5, 124.8, 123.24, 114.6, 48.97, 45.12.

MS (EI, 70 eV) *m/z* (%) = 248.9 (20) [M⁺], 247.9 (100), 144.9 (29), 142.9 (35), 91 (25).

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