Supplementary Information

Preparation of thermally curable materials using lignin extracted from sawmill co-product

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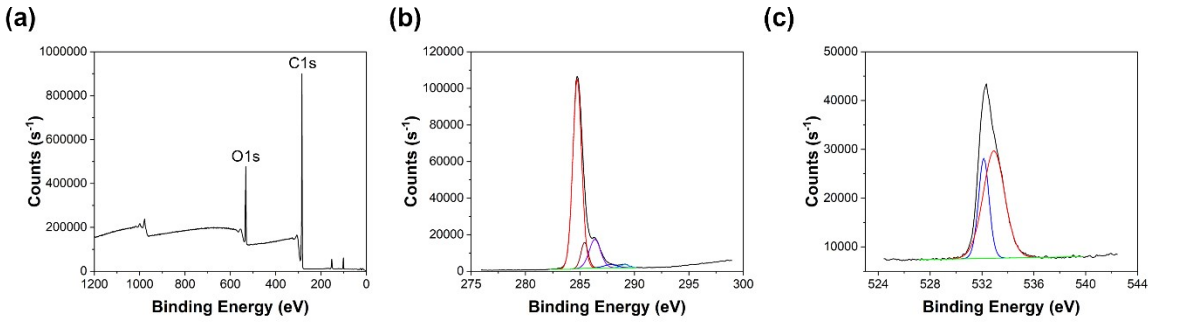
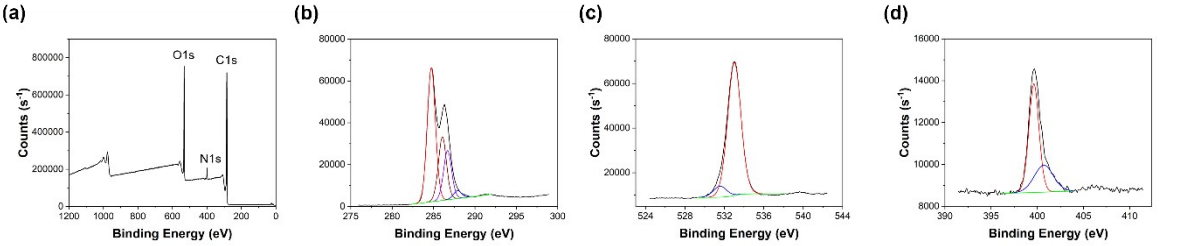


	BE range (eV)	Area (%)
C 1s	292.6 - 280.1	64.6
O 1s	539.1 – 527.0	28.1
N 1s	-	-

	BE (eV)	Area (%)
O-C=O	289.1	1.22
C=O	287.8	1.94
C-OH/C-O-C	286.4	14.4
C-C sp3	285.4	8.28
C-C sp2	284.8	74.2

	BE (eV)	Area (%)
C-OH/C-O-C	532.9	68.6
C=O	532.1	31.4

Figure S1. XPS spectra collected for unmodified Sonichem lignin: (a) survey spectrum; (b) C region with fits for C-C sp2 (red), C-C sp3 (dark red), C-OH/C-O-C (purple), O-C=O (light blue), C=O (blue); (c) O region with fits for C=O (blue) and C-OH/C-O-C (red). The background is shown in green. The tables underneath each graph summarise the results from fitting the data.



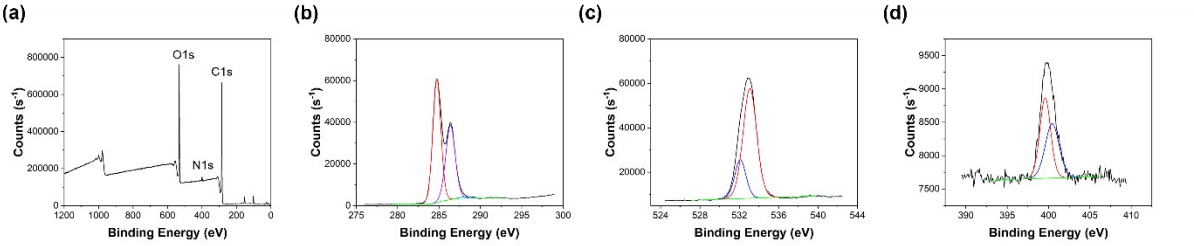
	BE range (eV)	Area (%)
C 1s	295.0 - 280.3	57.9
O 1s	542.3 – 527.0	38.5
N 1s	407.0 - 395.0	3.57

	BE (eV)	Area (%)
C=O	287.9	3.14
C-OH/C-O-C/ C-N	286.7	19.3
C-C sp3	285.9	24.9
C-C sp2	284.8	52.6

	BE (eV)	Area (%)
C-OH/C-O-C	533.1	92.3
C=O	531.6	7.72

	BE (eV)	Area (%)
N-sp2 C	400.7	32.1
N-sp3 C	399.6	67.9

Figure S2. XPS spectra collected for SL-BZ-aniline: (a) survey spectrum; (b) C region with fits for C-C sp2 (red), C-C sp3 (dark red), C-OH/C-O-C/C-N (purple), C=O (blue); (c) O region with fits for C=O (blue) and C-OH/C-O-C (red); (d) N region with fits for N-sp2 C (red) and N-sp3 C (blue).The background is shown in green. The tables underneath each graph summarise the results from fitting the data.



	BE range (eV)	Area (%)
C 1s	295.0 - 280.1	51.8
O 1s	541.5 – 527.0	41.5
N 1s	407.0 - 395.0	2.42

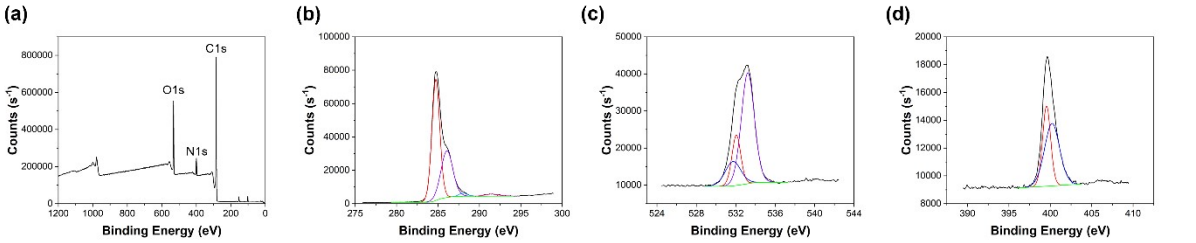
	BE (eV)	Area (%)
C=O	288.2	0.94
C-OH/C-O-C/ C-N	286.4	41.2
C-C	284.8	57.9

	BE (eV)	Area (%)
C-OH/C-O-C	533.1	77.5
C=O	532.0	22.5

	BE (eV)	Area (%)
N-sp2 C	400.4	46.6
N-sp3 C	399.6	53.4

Figure S3. XPS spectra collected for SL-BZ-A: (a) survey spectrum; (b) C region with fits for C-C sp2 and sp3 (red), C-OH/C-O-C/C-N (purple), C=O (blue); (c) O region with fits for C=O

(blue) and C-OH/C-O-C (red); (d) N region with fits for N-sp2 C (red) and N-sp3 C (blue).The background is shown in green. The tables underneath each graph summarise the results from



	BE range (eV)	Area (%)
C 1s	295.0 - 280.1	59.8
O 1s	539.1 – 527.0	29.1
N 1s	407.0 - 395.0	7.26

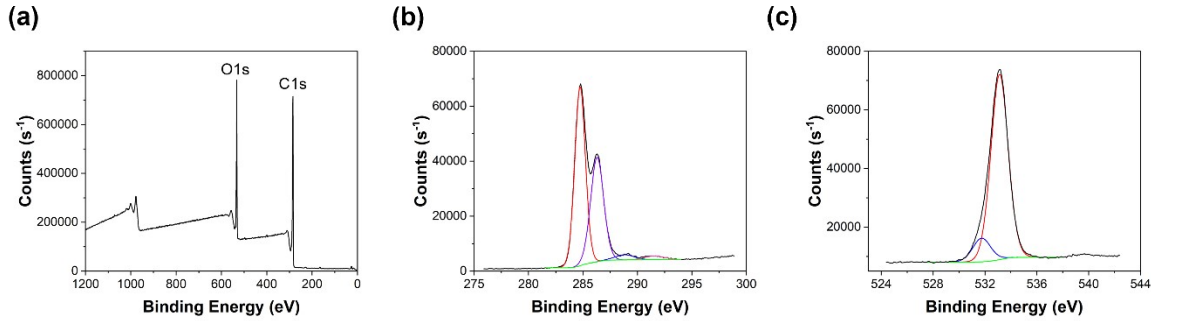
	BE (eV)	Area (%)
π-π* satellite	291.5	1.70
C=O	288.0	1.10
C-OH/C-O-C/ C-N	286.1	35.7
C-C	284.7	61.0

	BE (eV)	Area (%)
C-OH	533.2	63.8
C-O-C	532.1	19.5
C=O	531.7	16.7

	BE (eV)	Area (%)
N-sp2 C	400.2	59.4
N-sp3 C	399.6	40.6

fitting the data.

Figure S4. XPS spectra collected for SL-BZ-B: (a) survey spectrum; (b) C region with fits for C-C sp2 and sp3 (red), C-OH/C-O-C/C-N (purple), C=O (blue), π-π* satellites (pink); (c) O region with fits for C=O (blue), C-OH (purple) and C-O-C (red); (d) N region with fits for N-sp2 C (red) and N-sp3 C (blue). The background is shown in green. The tables underneath each graph summarise the results from fitting the data.

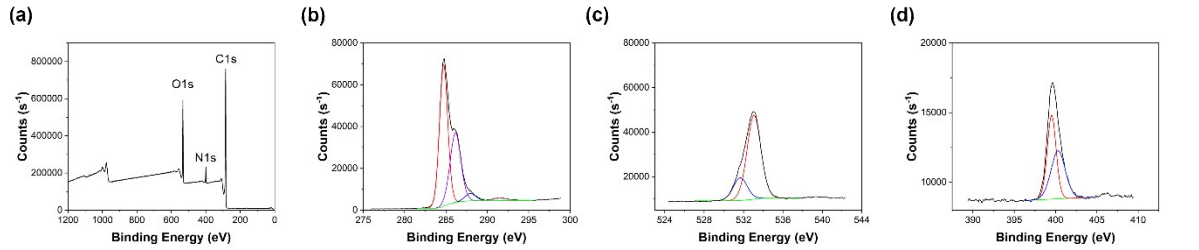


	BE range (eV)	Area (%)
C 1s	295.0 – 282.0	56.2
O 1s	541.7 – 528.1	43.8
N 1s	-	-

	BE (eV)	Area (%)
π-π* satellite	291.6	2.14
C=O	288.3	2.85
C-OH/C-O-C	286.3	39.7
C-C	284.7	55.3

	BE (eV)	Area (%)
C-O	533.1	88.2
C=O	531.8	11.8

Figure S5. XPS spectra collected for KL: (a) survey spectrum; (b) C region with fits for C-C sp2 and sp3 (red), C-OH/C-O-C/C-N (purple), C=O (blue), π-π* satellites (pink); (c) O region with fits for C=O (blue) and C-OH/C-O-C (red).The background is shown in green. The tables underneath each graph summarise the results from fitting the data.



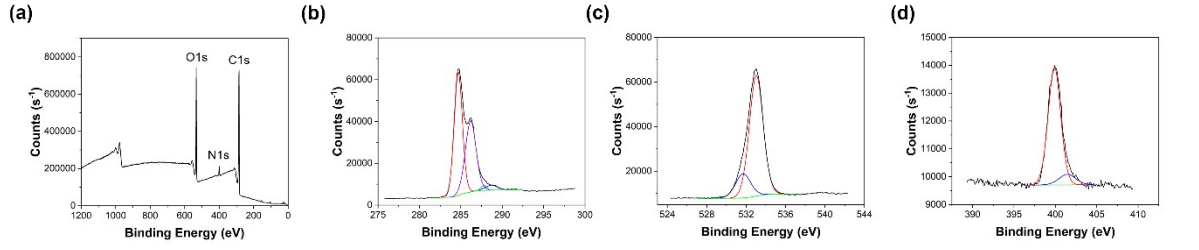
	BE range (eV)	Area (%)
C 1s	295.0 – 282.0	61.1
O 1s	541.7 – 528.0	33.3
N 1s	407.0 - 395.0	5.62

	BE (eV)	Area (%)
π-π* satellite	291.5	1.67
C=O	288.0	3.91
C-OH/C-O-C/ C-N	286.1	37.1
C-C	284.7	57.4

	BE (eV)	Area (%)
C-O	533.1	78.5
C=O	531.7	21.5

	BE (eV)	Area (%)
N-sp2 C	400.3	47.9
N-sp3 C	399.5	52.1

Figure S6. XPS spectra collected for KL-BZ-aniline: (a) survey spectrum; (b) C region with fits for C-C sp2 and sp3 (red), C-OH/C-O-C/C-N (purple), C=O (blue), π-π* satellites (pink); (c) O region with fits for C=O (blue), C-OH/C-O-C (red); (d) N region with fits for N-sp2 C (red) and N-sp3 C (blue). The background is shown in green. The tables underneath each graph summarise the results from fitting the data.



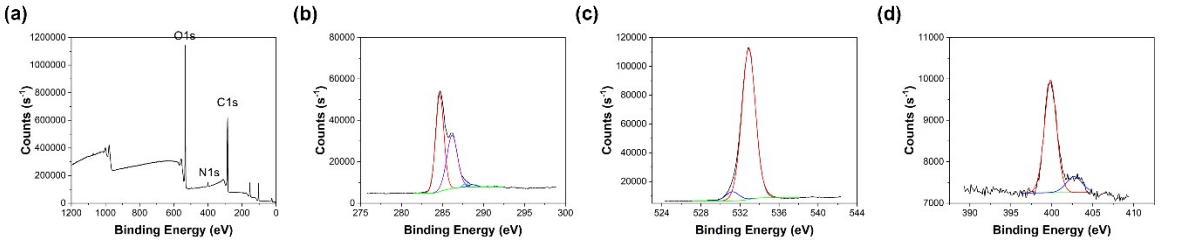
	BE range (eV)	Area (%)
C 1s	295.0 – 282.0	53.7
O 1s	541.7 – 528.1	41.7
N 1s	407.0 - 395.0	3.38

	BE (eV)	Area (%)
O-C=O	287.8	1.47
C=O	288.7	1.74
C-OH/C-O-C/ C-N	286.2	41.0
C-C	284.7	54.2

	BE (eV)	Area (%)
C-O	533.0	82.0
C=O	531.7	18.0

	BE (eV)	Area (%)
N-sp2 C	401.5	10.7
N-sp3 C	399.9	89.3

Figure S7. XPS spectra collected for KL-BZ-A: (a) survey spectrum; (b) C region with fits for C-C sp2 and sp3 (red), C-OH/C-O-C/C-N (purple), O-C=O (light blue), C=O (blue); (c) O region with fits for C=O (blue), C-OH/C-O-C (red); (d) N region with fits for N-sp2 C (red) and N-sp3 C (blue). The background is shown in green. The tables underneath each graph summarise the results from fitting the data.



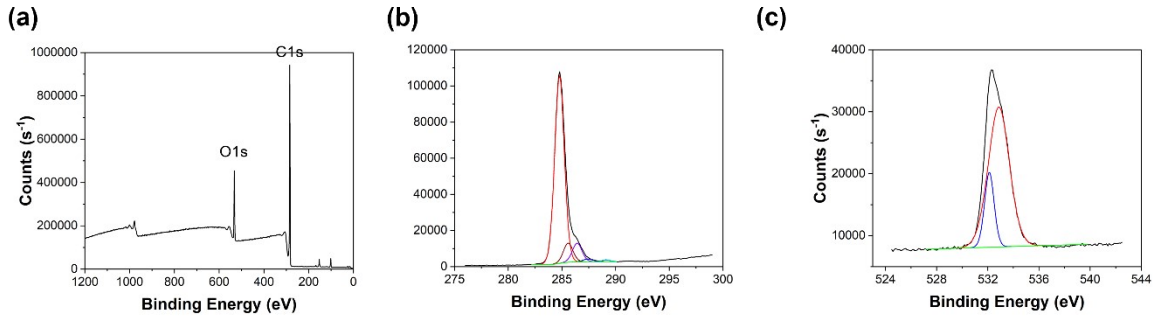
	BE range (eV)	Area (%)
C 1s	295.0 – 282.0	33.8
O 1s	541.7 – 528.0	52.4
N 1s	407.0 - 395.0	1.87

	BE (eV)	Area (%)
O-C=O	287.8	1.24
C=O	288.7	1.58
C-OH/C-O-C/ C-N	286.2	40.8
C-C	284.7	56.4

	BE (eV)	Area (%)
C-O	532.9	94.4
C=O	531.2	5.57

	BE (eV)	Area (%)
N-sp2 C	402.7	15.6
N-sp3 C	399.8	84.4

Figure S8. XPS spectra collected for KL-BZ-B: (a) survey spectrum; (b) C region with fits for C-C sp2 and sp3 (red), C-OH/C-O-C/C-N (purple), O-C=O (light blue), C=O (blue); (c) O region with fits for C=O (blue), C-OH/C-O-C (red); (d) N region with fits for N-sp2 C (red) and N-sp3 C (blue). The background is shown in green. The tables underneath each graph summarise the results from fitting the data.



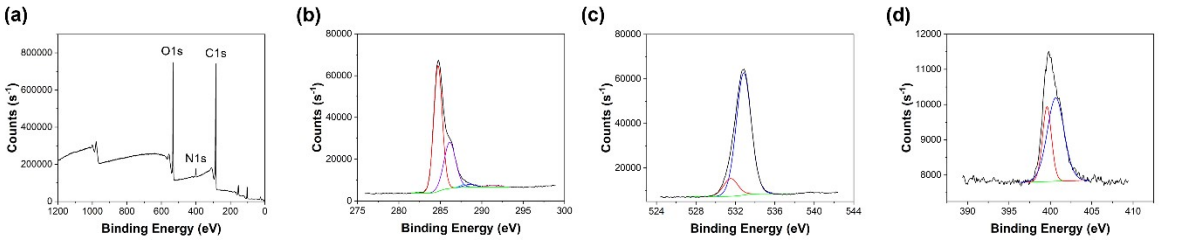
	BE range (eV)	Area (%)
C 1s	293.3 - 280.1	67.9
O 1s	539.1 – 527.0	25.9
N 1s	-	-

	BE (eV)	Area (%)
O-C=O	289.1	0.71
C=O	287.4	1.23
C-OH/C-O-C	286.4	8.16
C-C sp3	285.6	8.34
C-C sp2	284.7	81.6

	BE (eV)	Area (%)
C-OH/C-O-C	532.9	80.4
C=O	532.1	19.6

the results from fitting the data.

Figure S9. XPS spectra collected for SL-Ph: (a) survey spectrum; (b) C region with fits for C-C sp2 (red), C-C sp3 (dark red) C-OH/C-O-C (purple),O-C=O (light blue), C=O (blue); (c) O region with fits for C=O (blue) and C-OH/C-O-C (red). The tables underneath each graph summarise the results from fitting the data.



	BE range (eV)	Area (%)
C 1s	295.0 - 280.1	47.0
O 1s	541.7 – 528.1	40.6
N 1s	407.0 - 395.0	3.37

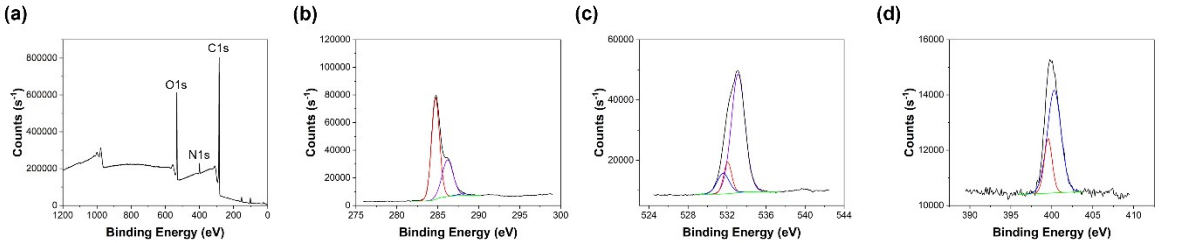
	BE (eV)	Area (%)
π-π* satellite	291.4	1.37
C=O	288.6	1.99
O-C-O	287.8	1.13
C-OH/C-O-C/ C-N	286.4	33.1
C-C	284.7	63.4

	BE (eV)	Area (%)
C-O	532.9	87.63
C=O	531.5	12.4

	BE (eV)	Area (%)
N-sp2 C	400.6	67.64
N-sp3 C	399.6	32.36

Figure S10. XPS spectra collected for SL-PhBZ-aniline: (a) survey spectrum; (b) C region with fits for C-C sp2 (red), C-C sp3 (dark red) C-OH/C-O-C/C-N (purple),O-C=O (light blue), C=O (blue), π-π* satellites (pink); (c) O region with fits for C=O (blue), C-OH/C-O-C (red); (d) N

region with fits for N-sp2 C (red) and N-sp3 C (blue). The tables underneath each graph summarise the results from fitting the data.



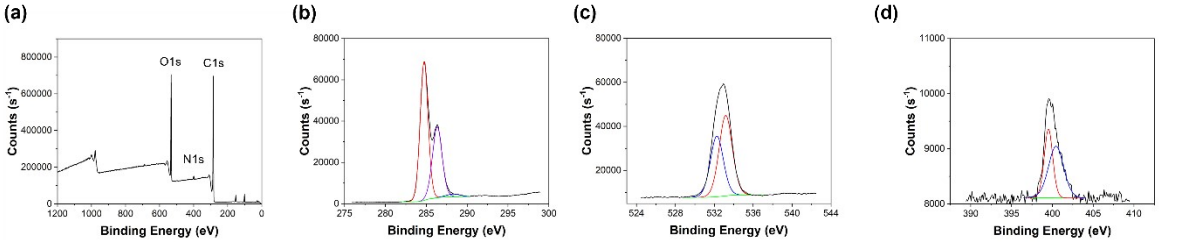
	BE range (eV)	Area (%)
C 1s	295.0 - 280.1	57.7
O 1s	541.4 – 527.0	34.7
N 1s	407.0 - 395.0	3.75

	BE (eV)	Area (%)
C=O	288.0	1.62
C-OH/C-O-C/ C-N	286.4	33.1
C-C	284.7	65.4

	BE (eV)	Area (%)
C-OH	533.1	76.4
C-O-C	532.1	11.9
C=O	531.6	11.8

	BE (eV)	Area (%)
N-sp2 C	400.3	76.3
N-sp3 C	399.6	23.7

Figure S11. XPS spectra collected for SL-PhBZ-A: (a) survey spectrum; (b) C region with fits for C-C sp2 and sp3 (red), C-OH/C-O-C/C-N (purple), C=O (blue); (c) O region with fits for C=O (blue), C-OH (purple) and C-O-C (red); (d) N region with fits for N-sp2 C (red) and N-sp3 C (blue). The tables underneath each graph summarise the results from fitting the data.



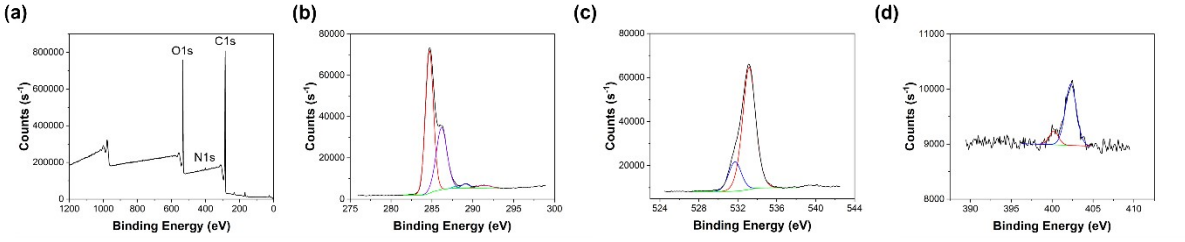
	BE range (eV)	Area (%)
C 1s	295.0 - 280.1	54.2
O 1s	539.1 – 527.0	38.9
N 1s	407.0 - 395.0	1.62

	BE (eV)	Area (%)
C=O	288.3	2.00
C-OH/C-O-C/ C-N	286.3	37.8
C-C	284.7	60.2

	BE (eV)	Area (%)
C-OH/C-O-C	533.2	57.3
C=O	532.3	42.7

	BE (eV)	Area (%)
N-sp2 C	400.5	57.3
N-sp3 C	399.5	42.7

Figure S12. XPS spectra collected for SL-PhBZ-B: (a) survey spectrum; (b) C region with fits for C-C sp2and sp3 (red), C-OH/C-O-C/C-N (purple), C=O (blue); (c) O region with fits for C=O (blue), C-O (red); (d) N region with fits for N-sp2 C (red) and N-sp3 C (blue). The background is shown in green. The tables underneath each graph summarise the results from fitting the data.



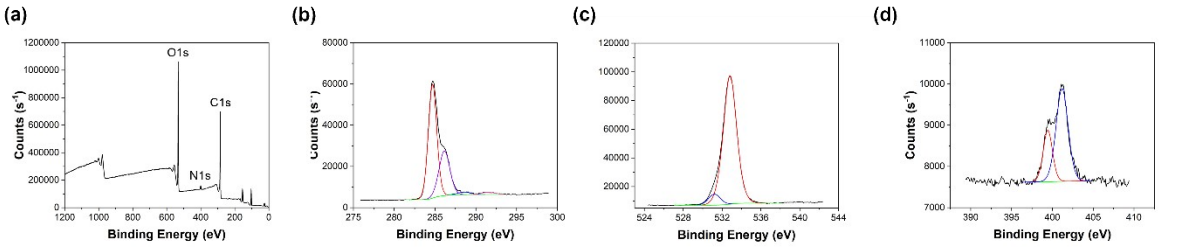
	BE range (eV)	Area (%)
C 1s	295.0 – 282.0	55.0
O 1s	541.7 – 528.1	41.2
N 1s	407.0 - 395.0	0.69

	BE (eV)	Area (%)
π-π* satellite	291.3	1.98
O-C=O	287.8	0.50
C=O	289.1	1.86
C-OH/C-O-C /C-N	286.2	35.8
C-C	284.7	59.8

	BE (eV)	Area (%)
C-O	533.2	81.2
C=O	531.7	18.8

	BE (eV)	Area (%)
N-sp2 C	402.3	84.3
N-sp3 C	400.2	15.7

Figure S13. XPS spectra collected for KL-Ph: (a) survey spectrum; (b) C region with fits for C-C sp2 and sp3 (red), C-OH/C-O-C/C-N (purple), O-C=O (light blue), C=O (blue), π-π* satellites (pink); (c) O region with fits for C=O (blue), C-OH/C-O-C (red); (d) N region with fits for N-sp2 C (red) and N-sp3 C (blue). The background is shown in green. The tables underneath each graph summarise the results from fitting the data.



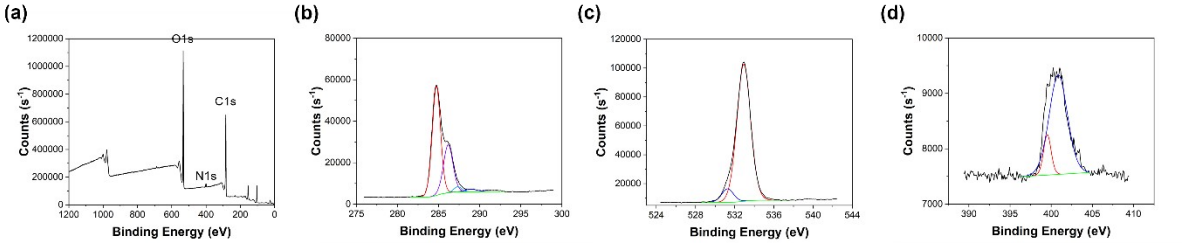
	BE range (eV)	Area (%)
C 1s	295.0 – 282.0	36.4
O 1s	541.7 – 528.1	48.7
N 1s	407.0 - 395.0	1.95

	BE (eV)	Area (%)
π-π* satellite	291.5	2.10
C=O	288.6	2.27
C-OH/C-O-C /C-N	286.2	32.2
C-C	284.7	63.5

	BE (eV)	Area (%)
C-O	532.8	93.0
C=O	531.2	7.01

	BE (eV)	Area (%)
N-sp2 C	401.2	70.2
N-sp3 C	399.5	29.8

Figure S14. XPS spectra collected for KL-PhBZ-aniline: (a) survey spectrum; (b) C region with fits for C-C sp2 and sp3 (red), C-OH/C-O-C/C-N (purple), C=O (blue), π-π* satellites (pink); (c) O region with fits for C=O (blue), C-OH/C-O-C (red); (d) N region with fits for N-sp2 C (red) and N-sp3 C (blue). The background is shown in green. The tables underneath each graph summarise the results from fitting the data.



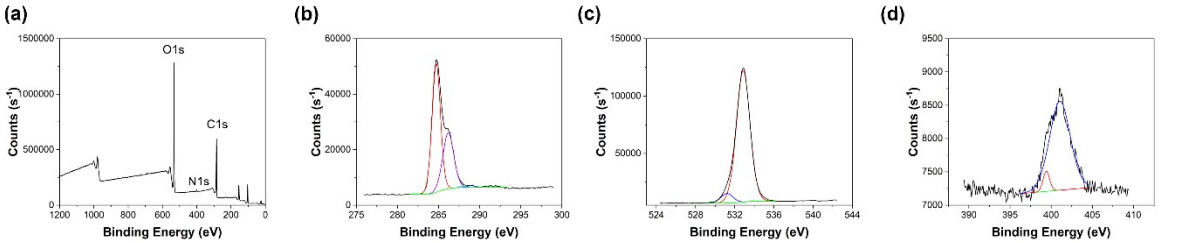
	BE range (eV)	Area (%)
C 1s	295.0 – 282.0	34.9
O 1s	541.7 – 528.1	51.5
N 1s	407.0 - 395.0	1.55

	BE (eV)	Area (%)
π-π* satellite	291.6	1.52
O-C=O	287.3	2.84
C=O	289.0	2.11
C-OH/C-O-C /C-N	286.2	31.5
C-C	284.7	62.0

	BE (eV)	Area (%)
C-O	532.9	92.1
C=O	531.3	7.9

	BE (eV)	Area (%)
N-sp2 C	400.9	85.6
N-sp3 C	399.5	14.4

Figure S15. XPS spectra collected for KL-PhBZ-A: (a) survey spectrum; (b) C region with fits for C-C sp2 and sp3 (red), C-OH/C-O-C/C-N (purple), O-C=O (light blue), C=O (blue), π-π* satellites (pink); (c) O region with fits for C=O (blue), C-OH/C-O-C (red); (d) N region with fits for N-sp2 C (red) and N-sp3 C (blue). The background is shown in green. The tables underneath each graph summarise the results from fitting the data.



	BE range (eV)	Area (%)
C 1s	295.0 – 282.0	28.5
O 1s	541.7 – 528.1	55.2
N 1s	407.0 - 395.0	1.32

	BE (eV)	Area (%)
π-π* satellite	291.4	1.14
O-C=O	287.9	0.47
C=O	288.9	0.75
C-OH/C-O-C /C-N	286.2	35.6
C-C	284.7	62.1

	BE (eV)	Area (%)
C-O	532.9	93.8
C=O	531.2	6.2

	BE (eV)	Area (%)
N-sp2 C	401.1	93.34
N-sp3 C	399.4	6.66

Figure S16. XPS spectra collected for KL-PhBZ-B: (a) survey spectrum; (b) C region with fits for C-C sp2 and sp3 (red), C-OH/C-O-C/C-N (purple), O-C=O (light blue), C=O (blue), π-π* satellites (pink); (c) O region with fits for C=O (blue), C-OH/C-O-C (red); (d) N region with fits for N-sp2 C (red) and N-sp3 C (blue). The background is shown in green. The tables underneath each graph summarise the results from fitting the data.

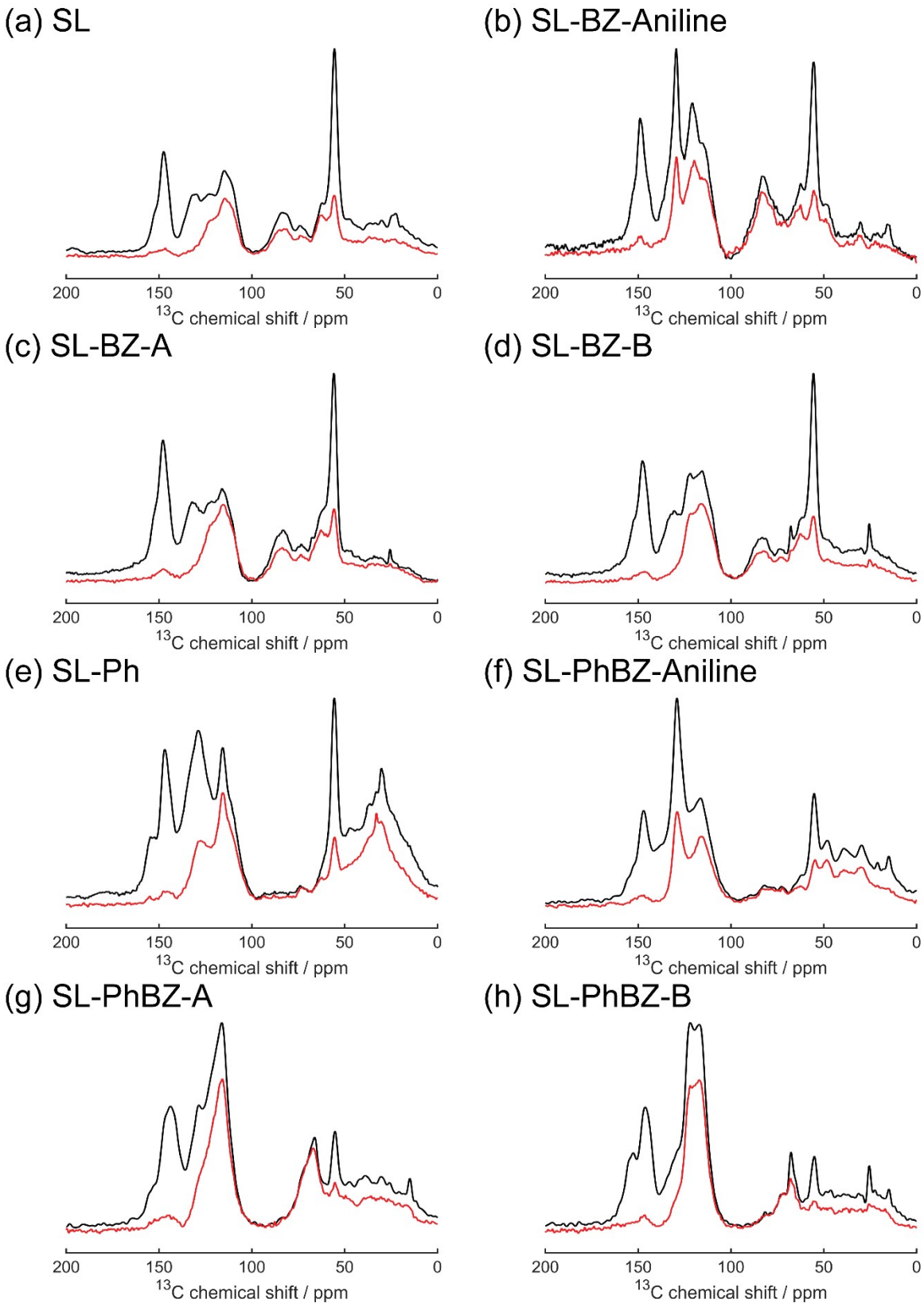


Figure S17. ^{13}C CP MAS NMR spectra of Sonichem lignin, functionalised with phenol and/or benzoxazine, detected with a CP contact time of 1 ms (black) and 50 μs (red).

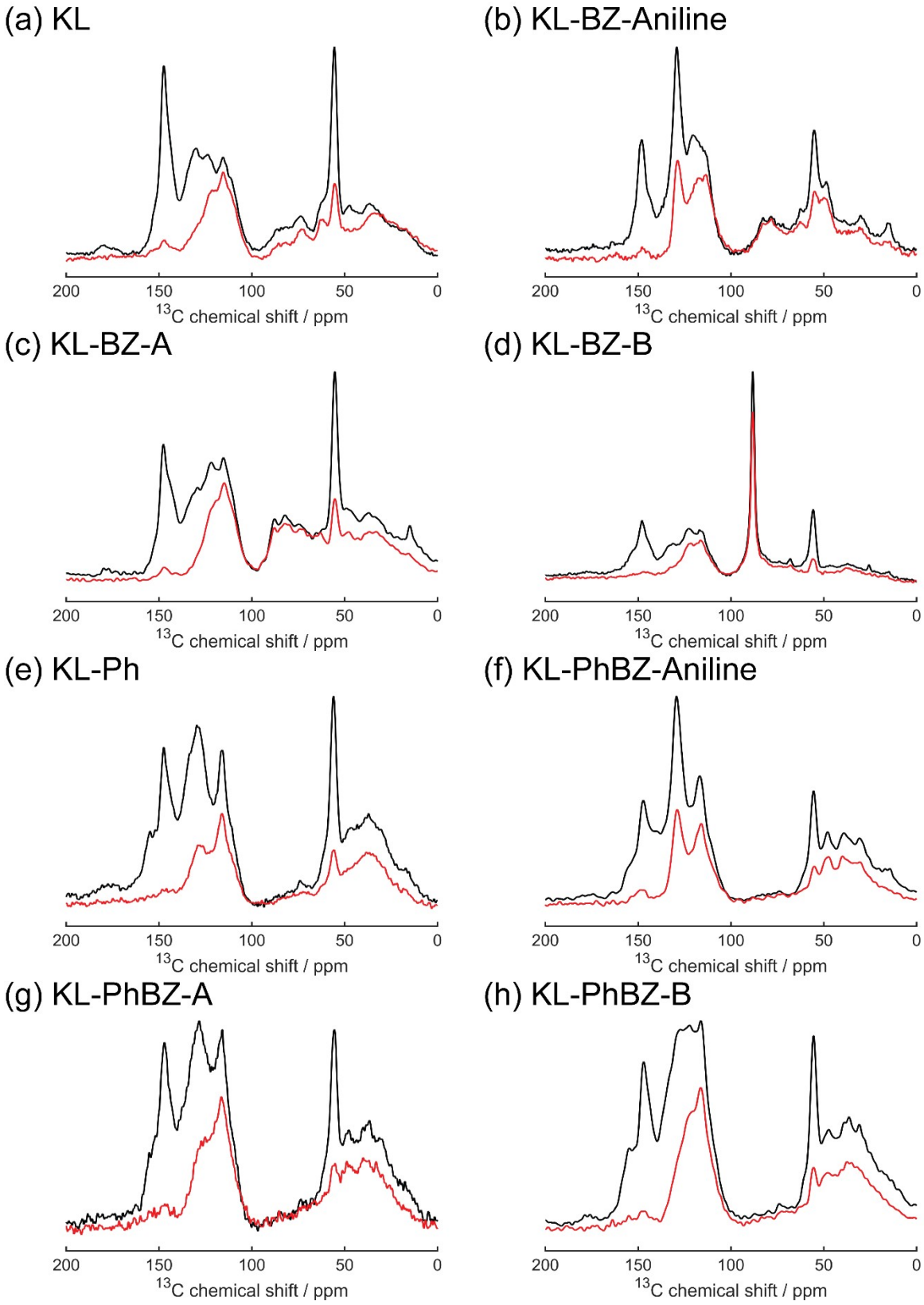


Figure S18. ^{13}C CP MAS NMR spectra of Kraft lignin, functionalised with phenol and/or benzoxazine, detected with a CP contact time of 1 ms (black) and 50 μs (red).

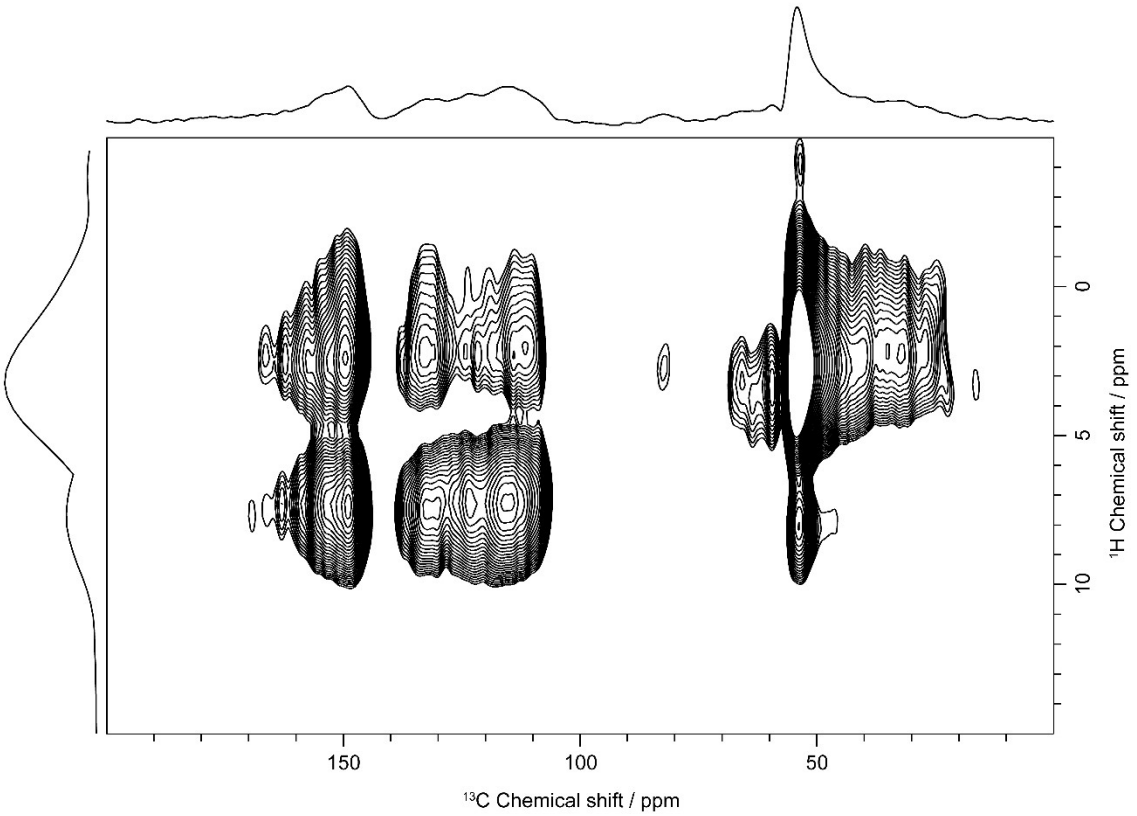


Figure S19: 2D ^{1}H–^{13}C HETCOR NMR spectrum of SL lignin, measured with a cross polarisation contact time of 1 ms.

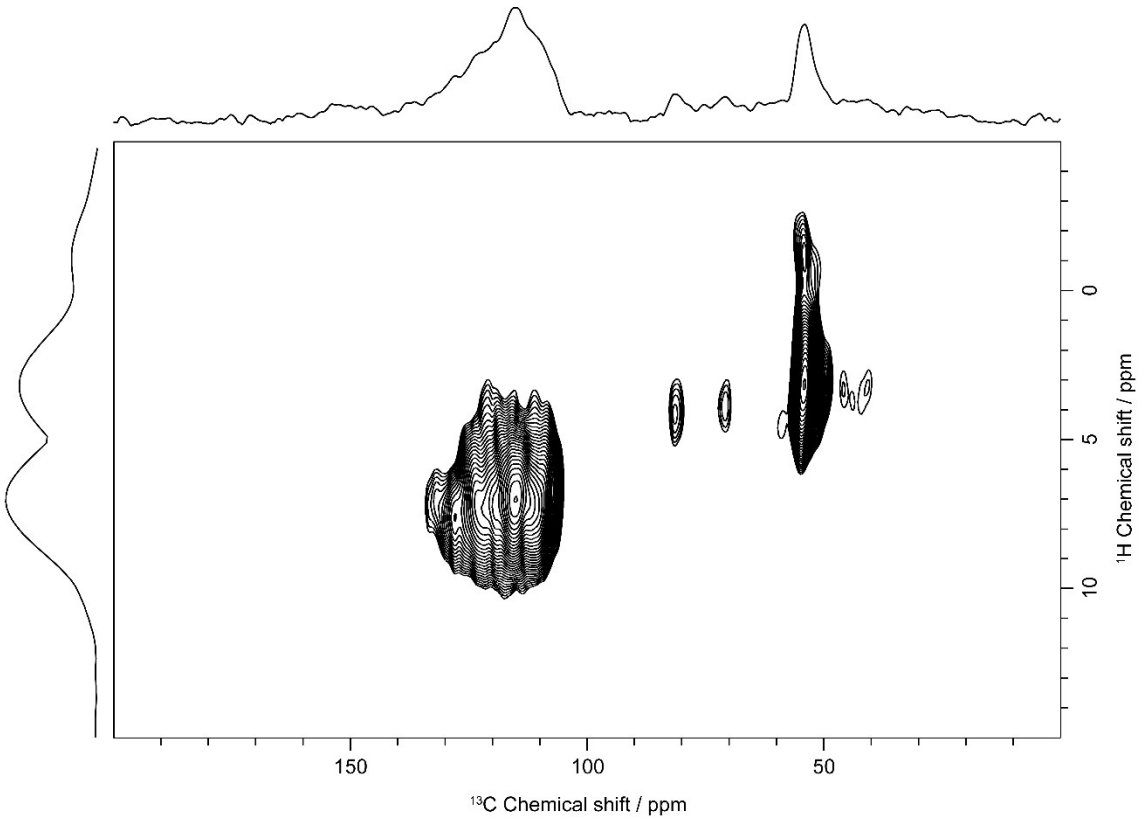


Figure S20: 2D ^{1}H–^{13}C HETCOR NMR spectrum of SL lignin, measured with a cross polarisation contact time of 50 µs.

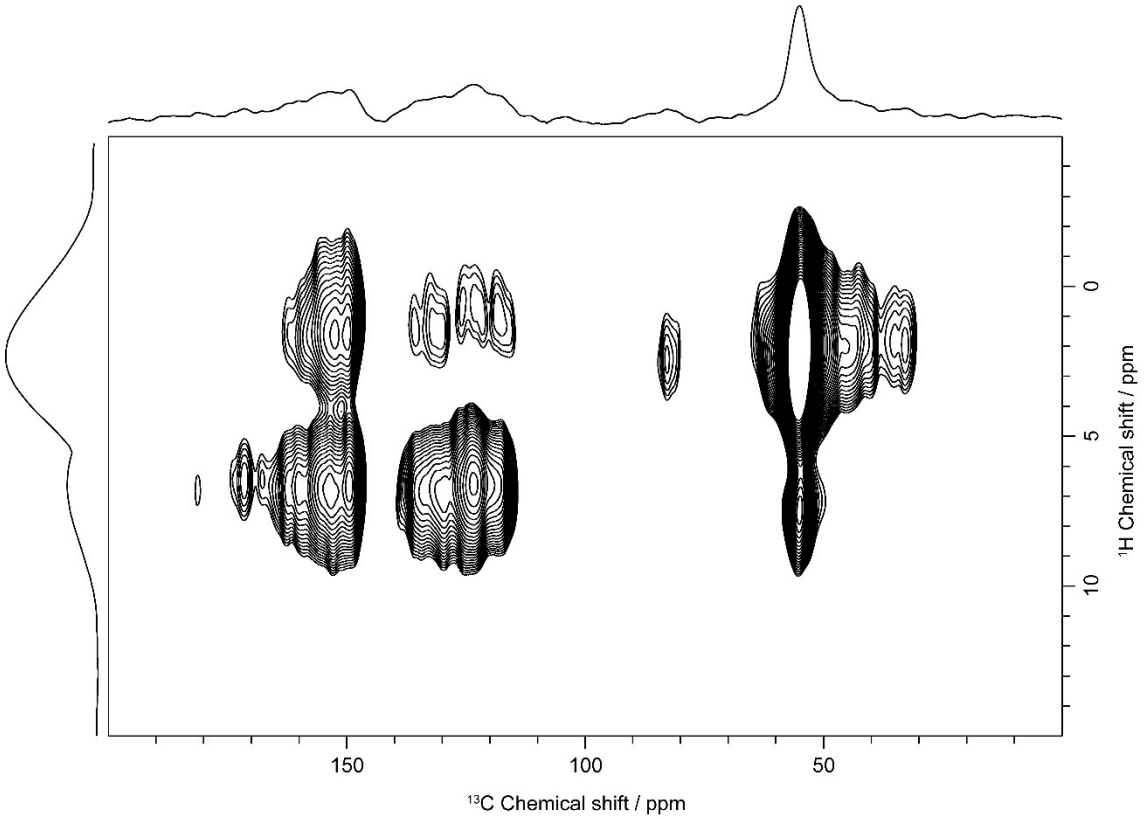


Figure S21: 2D ^{1}H–^{13}C HETCOR NMR spectrum of SL-BZ-B lignin, measured with a cross polarisation contact time of 1 ms.

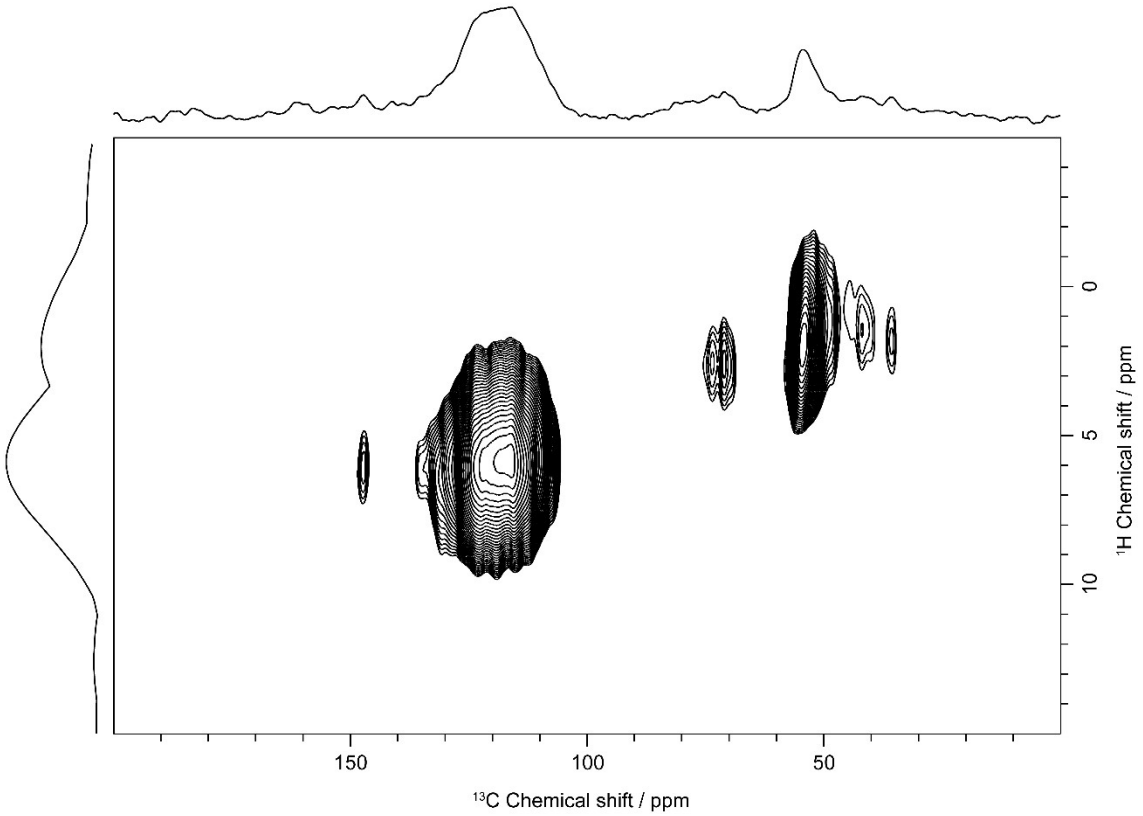


Figure S22: 2D ^{1}H–^{13}C HETCOR NMR spectrum of SL-BZ-B lignin, measured with a cross polarisation contact time of 50 µs.