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Sustainable polar aprotic/poly-deep eutectic solvent systems for highly efficient dissolution of lignin

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1. Table S1. The viscosity and density of PDESs at room temperature. 1
2. Fig. S1. The phase-transition temperatures of PDESs. 1
3. Fig. S2. Solubility of lignin in PDESs 1
4. Table S2. The lignin solubility in the pure PDESs and polar aprotic solvents at room temperature. 2
5. Table S3. The viscosity of PDESs with different co-solvents at room temperature. 2
6. Table S4. Solubility of EHL in the synthesized PDESs with different co-solvents at room temperature. 2
7. Table S5. Solubility of LS in the synthesized PDESs with different co-solvents at room temperature. 3
8. Table S6. Solubility of OL in the synthesized PDESs with different co-solvents at room temperature. 4
9. Fig. S3. 2D HSQC NMR spectra of the three lignin samples, including the side chain regions and the aromatic regions. Main substructures of lignin, including side chain linkages and aromatic units. 5
10. Table S7. Assignments of ^{13}C-^{1}H correlated signals in the HSQC spectra of three lignin samples. 5
11. Table S8. The Kamlet-Taft empirical parameters of solvent. 6
12. Table S9. The Kamlet-Taft empirical parameters of the PC/PDESs.a 6
13. Table S10. The Kamlet-Taft empirical parameters of the NFM/PDESs.[a] 7
14. Table S11. The Kamlet-Taft empirical parameters of the DMI/PDESs.[a] 8
15. Table S12. The Kamlet-Taft empirical parameters of the Cyrene/PDESs.a 9
16. Fig. S4. The correlation between the LS solubility and the α value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents). 10

17. Fig. S5. The correlation between the OL solubility and the α value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents). 10
18. Fig. S6. The correlation between the EHL solubility and the β value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents). 11
19. Fig. S7. The correlation between the LS solubility and the β value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents). 11
20. Fig. S8. The correlation between the OL solubility and the β value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents). 12
21. Fig. S9. The correlation between the EHL solubility and the α value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents). 12
22. Fig. S10. The correlation between the EHL solubility and the π* value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents, and the critical value of π* was 0.441, 0.452, 0.587, 0.520, 0.418 and 0.554 in P2-Lev, P4-Lev, P6-Lev, PA2-Lev, PA4-Lev, and PA6-Lev, respectively). 13
23. Fig. S11. The correlation between the OL solubility and the π* value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents, and the critical value of π* was 0.441, 0.452, 0.531, 0.441, 0.418 and 0.406 in P2-Lev, P4-Lev, P6-Lev, PA2-Lev, PA4-Lev, and PA6-Lev, respectively). 13
24. Fig. S12. SEM images of a), b) and c) initial LS and d), e) and f) regenerated LS. 14
25. Fig. S13. SEM images of a), b) and c) initial OL and d), e) and f) regenerated OL. 14
26. Fig. S14. SEM images of a), b) and c) initial EHL and d), e) and f) regenerated EHL. 14
27. Fig. S15. The FT-IR of native and generated lignins 15
28. Fig. S16. TGA of initial and regenerated lignin 15
29. Table S13. Sugar content and molecular weight of three lignin samples. 15
30. Table S14. Element analysis of different lignin 16

Table S1. The viscosity and density of PDESs at room temperature.

Entry	PDES	Viscosity (mPa·s)	Density (g/cm^{-3})
1	P2-Lev	51.20	1.1338
2	P4-Lev	94.72	1.1331
3	P6-Lev	133.26	1.1314
4	PA2-Lev	29.40	1.1280
5	PA4-Lev	58.84	1.1278
6	PA6-Lev	96.90	1.1271

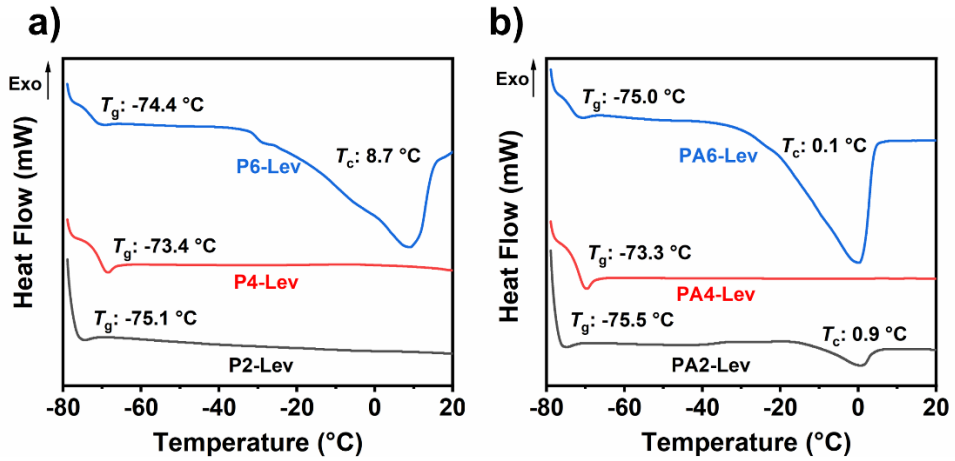

Fig. S1. The phase-transition temperatures of PDESs.

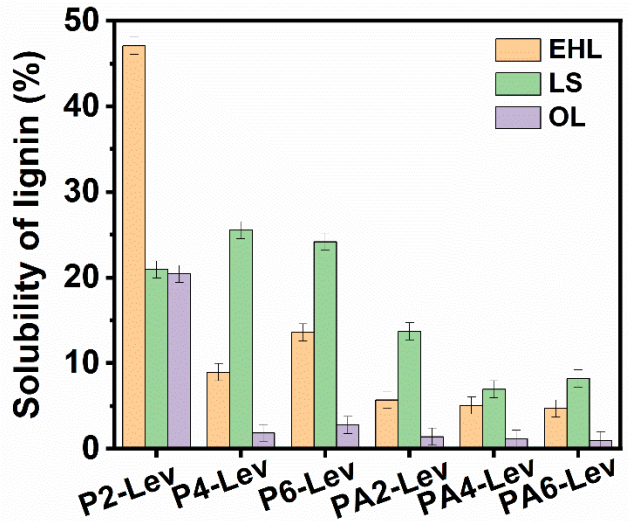

Fig. S2. Solubility of lignin in PDESs

Table S2. The lignin solubility in the pure PDESs and polar aprotic solvents at room temperature.

Entry	Solvent	Solubility (%)		
		EHL	OL	LS
1	P2-Lev	47.06 (±1.2)	20.41 (±1.2)	20.94 (±1.2)
2	P4-Lev	8.94 (±0.1)	1.82 (±0.1)	25.52 (±0.2)
3	P6-Lev	13.5 (±0.4)	2.76 (±0.1)	24.17 (±0.5)
4	PA2-Lev	5.71 (±0.4)	1.41 (±0.5)	13.70 (±1.0)
5	PA4-Lev	5.06 (±0.5)	1.17 (±0.2)	6.94 (±0.5)
6	PA6-Lev	4.71 (±0.1)	0.94 (±0.1)	8.20 (±0.4)
7	PC	0.76 (±0.1)	0.82 (±0.1)	0.59 (±0.1)
8	NFM	0.58 (±0.1)	0.96 (±0.1)	0.59 (±0.1)
9	DMI	1.29 (±0.1)	0.59 (±0.1)	0.59 (±0.1)
10	Cyrene	0.58 (±0.1)	1.47 (±0.1)	0.59 (±0.1)

[a]The solubility value was the average of twice measurements, and the values in the parentheses were the half of the range of replicate results.

Table S3. The viscosity of PDESs with different co-solvents at room temperature.

Entry	PDES	Co-solvent (20 vol%)	Viscosity (mPa·s)
1	P6-Lev	DMI	87.58
2		PC	54.40
3		NFM	86.86
4		Cyrene	106.58

Table S4. Solubility of EHL in the synthesized PDESs with different co-solvents at room temperature.

Entry	PDES	Co-solvent content (vol%)	Solubility of EHL (%)[a]			
			PC	NFM	DMI	Cyrene
1	P2-Lev	10	62.00 (±1.7)	63.12 (±1.5)	70.00 (±1.7)	12.01 (±1.5)
2		20	72.47 (±1.8)	73.17 (±1.2)	80.23 (±1.7)	24.64 (±1.2)
3		40	72.52 (±1.5)	76.47 (±1.7)	67.35 (±1.4)	24.23 (±0.6)
4		60	60.41 (±1.7)	76.17 (±1.2)	60.47 (±1.7)	13.23 (±1.2)
5		80	54.29 (±1.1)	69.47 (±1.7)	55.35 (±0.7)	7.71 (±0.6)
6	P4-Lev	10	35.75 (±1.7)	30.14 (±1.6)	28.79 (±1.5)	10.44 (±1.6)
7		20	55.76 (±1.7)	47.17 (±1.7)	43.82 (±1.0)	15.47 (±1.0)
8		40	43.17 (±1.1)	37.47 (±1.1)	33.88 (±1.0)	14.23 (±0.5)
9		60	37.35 (±1.1)	30.11 (±1.0)	27.11 (±0.5)	8.12 (±1.0)
10		80	25.41 (±0.5)	21.29 (±0.6)	17.17 (±0.5)	6.24 (±0.5)
11	P6-Lev	10	17.29 (±0.5)	10.58 (±0.4)	11.64 (±0.6)	12.8 (±0.5)
12		20	25.29 (±0.5)	11.58 (±0.7)	12.64 (±0.6)	13.47 (±0.5)
13		40	19.23 (±0.5)	9.76 (±0.7)	9.12 (±0.7)	12.47 (±0.5)
14		60	13.38 (±0.6)	6.71 (±0.6)	5.06 (±0.6)	7.88 (±0.6)
15		80	9.12 (±0.5)	3.35 (±0.5)	2.35 (±0.5)	4.00 (±0.5)
16	PA2-Lev	10	17.7 (±0.4)	8.141 (±0.5)	6.92 (±0.5)	6.03 (±0.4)
17		20	24.70 (±0.4)	10.41 (±0.5)	9.82 (±0.5)	8.53 (±0.3)

18		40	21.29 (±0.6)	8.00 (±0.6)	7.59 (±0.6)	8.18 (±0.6)
19		60	18.94 (±0.4)	4.76 (±0.4)	5.75 (±0.4)	7.71 (±0.4)
20		80	17.11 (±0. 6)	2.41 (±0.3)	2.53 (±0.3)	2.29 (±0.3)
21		10	28.21 (±0.5)	9.70 (±1.1)	15.01 (±0.8)	8.75 (±0.5)
22		20	32.47 (±1.0)	11.52 (±1.0)	19.05 (±0.8)	13.52 (±0.5)
23	PA4-Lev	40	21.35 (±0.8)	10.41 (±0.5)	14.00 (±0.5)	6.47 (±0.6)
24		60	9.41 (±0.5)	7.12 (±0.5)	9.94 (±0.4)	4.67 (±0.5)
25		80	7.12 (±0.5)	5.941 (±0.4)	7.82 (±0.5)	2.18 (±0.4)
26		10	20.55 (±0.6)	5.23 (±0.5)	3.75 (±0.4)	5.1 (±0.3)
27		20	26.94 (±0.5)	7.53 (±0.4)	4.12 (±0.4)	5.59 (±0.4)
28	PA6-Lev	40	23.17 (±0.8)	4.12 (±0.4)	2.24 (±0.3)	5.16 (±0.4)
29		60	20.17 (±0.5)	2.12 (±0.5)	2.00 (±0.6)	4.24 (±0.5)
30		80	15.11 (±0.2)	1.29 (±0.3)	1.71 (±0.5)	1.88 (±0.3)

[a]The solubility value was the average of twice measurements, and the values in the parentheses were the half of the range of replicate results

Table S5. Solubility of LS in the synthesized PDESs with different co-solvents at room temperature.

Entry	**PDES**	**Co-solvent content (vol%)**	**Solubility of LS (%)[a]**			
			PC	**NFM**	**DMI**	**Cyrene**
1		10	82.45 (±1.6)	68.89 (±1.4)	78.83 (±1.5)	38.20 (±1.7)
2		20	92.52 (±1.7)	78.94 (±1.7)	96.82 (±1.7)	48.23 (±1.1)
3	P2-Lev	40	60.52 (±1.7)	56.05 (±1.1)	63.00 (±1.7)	45.82 (±1.1)
4		60	54.23 (±1.1)	36.17 (±0.8)	54.94 (±1.1)	29.35 (±1)
5		80	48.35 (±.1)	21.17 (±0.8)	46.47 (±1.1)	11.52 (±1.1)
6		10	42.31 (±1.4)	42.60 (±1.6)	47.8 (±1.6)	31.8 (±1.7)
7		20	54.35 (±1.7)	53.64 (±1.7)	59.82 (±1.1)	40.82 (±1.7)
8	P4-Lev	40	44.64 (±1.1)	44.64 (±1.1)	51.82 (±1.7)	34.76 (±1.7)
9		60	31.71 (±1.1)	33.88 (±1.1)	41.05 (±1.7)	18.71 (±1.1)
10		80	26.17 (±0.5)	18.00 (±1.1)	20.11 (±1.1)	8.18 (±0.5)
11		10	25.13 (±1.0)	20.15 (±0.6)	20.23 (±1.2)	30.12 (±1.1)
12		20	31.82 (±1.7)	24.14 (±1.1)	23.82 (±0.5)	33.35 (±1.7)
13	P6-Lev	40	24.71 (±1.2)	22.52 (±1.2)	17.64 (±1.2)	22.35 (±1.0)
14		60	15.82 (±1.2)	14.94 (±1.2)	12.52 (±1.0)	14.17 (±1.1)
15		80	9.18 (±0.5)	7.35 (±0.5)	4.529 (±0.5)	6.59 (±0.5)
16		10	44.2 (±2.0)	25.23 (±2.0)	30.1 (±1.8)	30.1 (±2.0)
17		20	52.47 (±2.0)	35.47 (±1.8)	41.76 (±2.0)	37.64 (±2.0)
18	PA2-Lev	40	48.11 (±1.2)	31.05 (±1.2)	29.41 (±1.0)	36.05 (±1.4)
19		60	35.52 (±1.0)	15.88 (±1.4)	14.64 (±1.2)	27.23 (±1.0)
20		80	29.82 (±1.0)	10.82 (±1.0)	4.24 (±1.0)	13.88 (±1.0)
21		10	22.2 (±1.2)	16.87 (±1.2)	23.60 (±1.1)	33.40 (±1.0)
22		20	26.58 (±1.2)	20.88 (±1.2)	29.41 (±1.2)	34.94 (±0.7)
23	PA4-Lev	40	16.29 (±1.0)	19.41 (±0.6)	16.52 (±1.1)	25.41 (±1.1)
24		60	9.88 (±0.6)	12.05 (±0.5)	12.94 (±0.5)	16.71 (±0.5)
25		80	7.05 (±0.5)	7.24 (±0.5)	9.41 (±0.6)	7.50 (±0.5)

26		10	16.21 (±0.4)	10.99 (±0.6)	9.76 (±0.6)	20.12 (±0.5)
27		20	19.00 (±0.4)	13.94 (±0.6)	12.11 (±0.6)	23.41 (±0.5)
28	PA6-Lev	40	16.88 (±0.4)	13.23 (±0.4)	9.65 (±0.5)	18.35 (±0.4)
29		60	12.17 (±0.3)	8.29 (±0.3)	7.71 (±0.3)	12.88 (±0.3)
30		80	9.05 (±0.3)	5.88 (±0.3)	5.24 (±0.3)	5.06 (±0.3)

[a]The solubility value was the average of twice measurements, and the values in the parentheses were the half of the range of replicate results

Table S6. Solubility of OL in the synthesized PDESs with different co-solvents at room temperature.

Entry	PDES	Co-solvent content (vol%)	Solubility of OL (%)[a]			
			PC	NFM	DMI	Cyrene
1		10	46.34 (±1.4)	48.76 (±1.6)	42.71 (±1.6)	4.01 (±1.7)
2		20	56.41 (±1.7)	61.76 (±1.7)	52.70 (±1.6)	4.41 (±1.1)
3	P2-Lev	40	46.35 (±1.7)	50.05 (±1.6)	49.47 (±1.7)	4.17 (±1.7)
4		60	31.58 (±1.1)	41.23 (±1.0)	41.11 (±1.1)	3.70 (±1.7)
5		80	21.76 (±1.1)	31.82 (±1.1)	34.05 (±1.1)	3.35 (±1.1)
6		10	2.29 (±0.11)	2.05 (±0.10)	3.05 (±0.11)	2.9 (±0.10)
7		20	3.29 (±0.11)	3.06 (±0.1)	4.52 (±0.15)	4.23 (±0.1)
8	P4-Lev	40	2.41 (±0.1)	2.76 (±0.12)	4.24 (±0.17)	3.29 (±0.11)
9		60	2.12 (±0.12)	2.53 (±0.21)	3.94 (±0.16)	2.94 (±0.11)
10		80	1.89 (±0.1)	2.18 (±0.1)	3.53 (±0.15)	2.47 (±0.1)
11		10	2.91 (±0.14)	2.78 (±0.11)	3.23 (±0.10)	3.12 (±0.12)
12		20	3.29 (±0.1)	2.94 (±0.1)	3.82 (±0.1)	3.58 (±0.1)
13	P6-Lev	40	2.71 (±0.1)	2.06 (±0.1)	2.29 (±0.1)	2.82 (±0.1)
14		60	2.24 (±0.1)	1.76 (±0.1)	1.59 (±0.1)	2.35 (±0.1)
15		80	1.24 (±0.1)	1.47 (±0.1)	0.71 (±0.1)	2.00 (±0.1)
16		10	6.23 (±0.3)	2.15 (±0.3)	7.52 (±0.4)	2.1 (±0.4)
17		20	9.35 (±0.3)	3.59 (±0.3)	10.59 (±0.4)	3.82 (±0.4)
18	PA2-Lev	40	8.53 (±0.3)	3.24 (±0.4)	7.65 (±0.3)	2.88 (±0.2)
19		60	8.00 (±0.2)	2.94 (±0.3)	5.88 (±0.2)	2.18 (±0.2)
20		80	6.94 (±0.1)	2.65 (±0.1)	4.71 (±0.1)	1.65 (±0.3)
21		10	2 (±0.3)	3.76 (±0.3)	10.12 (±0.4)	1.78 (±0.3)
22		20	2.65 (±0.3)	4.18 (±0.3)	13.64 (±0.4)	2.82 (±0.3)
23	PA4-Lev	40	2.35 (±0.2)	4.00 (±0.3)	13.05 (±0.3)	2.35 (±0.2)
24		60	1.53 (±0.2)	3.53 (±0.2)	12.17 (±0.2)	2.11 (±0.2)
25		80	1.29 (±0.2)	3.35 (±0.2)	10.05 (±0.2)	1.76 (±0.2)
26		10	1.43 (±0.1)	2.1 (±0.1)	4.12 (±0.1)	1.89 (±0.1)
27		20	1.94 (±0.1)	3.06 (±0.1)	5.29 (±0.1)	2.52 (±0.1)
28	PA6-Lev	40	1.59 (±0.06)	2.71 (±0.1)	4.82 (±0.08)	2.11 (±0.08)
29		60	1.30 (±0.1)	2.35 (±0.06)	3.59 (±0.1)	1.35 (±0.06)
30		80	1.00 (±0.08)	2.06 (±0.1)	2.76 (±0.1)	0.70 (±0.1)

[a]The solubility value was the average of twice measurements, and the values in the parentheses were the half of the range of replicate results

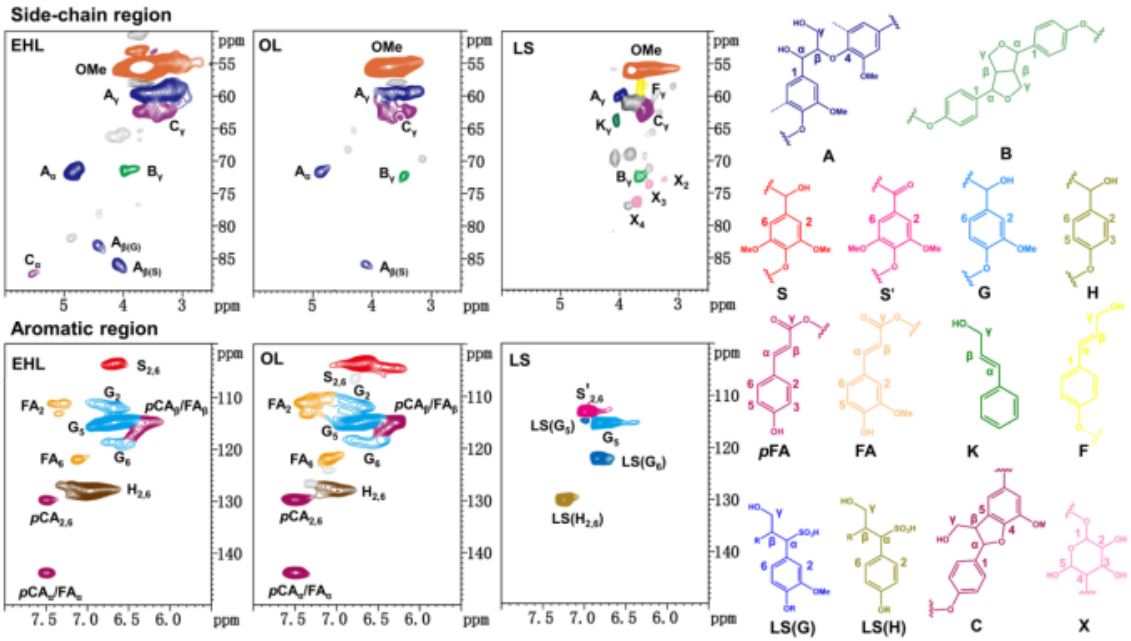

Fig. S3. 2D HSQC NMR spectra of the three lignin samples, including the side chain regions and the aromatic regions. Main substructures of lignin, including side chain linkages and aromatic units.

Table S7. Assignments of ^{13}C-^{1}H correlated signals in the HSQC spectra of three lignin samples.

Region	δC/δH (ppm)	Lable	Assignment
Aliphatic	55.7/3.73	OMe	C-H in methoxyls
	71.9/4.86	Aα	Cα-Hα in β-O-4' substructures (A)
	84.2/4.38	Aβ(G)	Cβ-Hβ in β-O-4' substructures to G (A)
	86.0/4.12	Aβ(S)	Cβ-Hβ in β-O-4' substructures to S (A)
	59.8/3.38-3.80	Aγ	Cγ-Hγ in β-O-4' substructures (A)
	72.2/3.72-4.15	Bγ	Cγ-Hγ in β-β resinol (B)
	87.7/5.54	Cα	Cα-Hα in phenylcoumaran (C)
	62.1/3.69	Cγ	Cγ-Hγ in phenylcoumaran (C)
	63.8/4.10	Kγ	Cγ-Hγ in cinnamyl alcohol (K)
	58.9/3.70	Fγ	Cγ-Hγ in p-hydroxycinnamyl alcohol (F)
	72.8/3.34	X2	C2-H2 in β-D-xylopyranoside (X)
	73.5/3.65	X3	C3-H3 in β-D-xylopyranoside (X)
	76.3/3.78	X4	C4-H4 in β-D-xylopyranoside (X)
Aromatic	104.0/6.71	S2,6	C2,6-H2,6 in syringyl units (S)
	104.5/7.34	S'2,6	C2,6-H2,6 in oxidized S units (S')
	110.9/6.99	G2	C2-H2 in guaiacyl units (G)
	115.6/6.78	G5	C5-H5 in guaiacyl units (G)
	115.5/6.97	LS(G_5)	C5-H5 in G units of lignosulfonate (LS)
	119.0/6.77	G6	C6-H6 in guaiacyl units (G)
	122.0/6.81	LS(G_6)	C6-H6 in G units of lignosulfonate (LS)
	127.5/7.05	H2,6	C2,6-H2,6 in p-hydroxyphenyl units (H)
	129.8/7.28	LS(H2,6)	C2,6-H2,6 in H units of lignosulfonate (LS)

110.1/7.29	FA_2	C_2-H_2 in ferulate (FA)
122.2/7.12	FA_6	C_6-H_6 in ferulate (FA)
130.1/7.51	$PCA_{2,6}$	$C_{2,6}$-$H_{2,6}$ in p-coumarate (PCA)
144.1/7.50	PCAα/FAα	Cα-Hα in p-coumarate (PCA) and ferulate (FA)
115.2/6.30	PCAβ/FAβ	Cβ-Hβ in p-coumarate (PCA) and ferulate (FA)

Table S8. The Kamlet-Taft empirical parameters of solvent.

Entry	Solvent	π*	α	β
1	P2-Lev	0.705	0.941	0.691
2	P4-Lev	0.630	0.850	0.801
3	P6-Lev	0.598	0.810	0.864
4	PA2-Lev	0.985	1.284	0.376
5	PA4-Lev	0.908	1.190	0.489
6	PA6-Lev	0.576	0.784	-0.201
7	DMI	0.441	0.619	0.909
8	PC	0.476	0.661	0.949
9	NFM	0.441	0.619	1.266
10	Cyrene	0.498	0.688	0.872

[a]The Kamlet-Taft empirical parameters was the average of twice measurements, and the values in the parentheses were the half of the range of replicate results

Table S9. The Kamlet-Taft empirical parameters of the PC/PDESs.a

Entry	PDES	PC content (vol%)	π*	α	β
1	P2-Lev	10	0.736	0.980	0.656
2		20	0.705	0.941	0.691
3		40	0.673	0.902	0.701
4		60	0.620	0.837	0.787
5		80	0.598	0.810	0.812
6	P4-Lev	10	0.620	0.837	1.114
7		20	0.620	0.837	1.041
8		40	0.600	0.837	0.966
9		60	0.576	0.784	0.965
10		80	0.521	0.716	0.950
11	P6-Lev	10	0.620	0.837	0.916
12		20	0.609	0.824	0.928
13		40	0.589	0.824	0.928
14		60	0.565	0.770	0.952
15		80	0.532	0.729	0.912
16	PA2-Lev	10	0.778	1.030	0.584
17		20	0.736	0.980	0.630
18		40	0.694	0.928	0.677
19		60	0.652	0.876	0.777

20		80	0.587	0.797	0.824
21		10	1.425	1.822	-0.067
22		20	1.417	1.812	-0.058
23	PA4-Lev	40	1.401	1.792	-0.040
24		60	1.392	1.782	-0.082
25		80	0.554	0.757	0.861
26		10	0.609	0.824	0.851
27		20	0.598	0.810	0.864
28	PA6-Lev	40	0.598	0.810	0.889
29		60	0.576	0.784	0.888
30		80	0.554	0.757	0.887

[a]The Kamlet-Taft empirical parameters was the average of twice measurements, and the values in the parentheses were the half of the range of replicate results

Table S10. The Kamlet-Taft empirical parameters of the NFM/PDESs.[a]

Entry	**PDES**	**NFM content (vol%)**	**π***	**α**	**β**
1		10	0.630	0.850	0.853
2		20	0.640	0.824	0.877
3	P2-Lev	40	0.587	0.797	0.901
4		60	0.521	0.716	1.052
5		80	0.499	0.702	1.140
6		10	0.609	0.824	0.877
7		20	0.576	0.784	0.965
8	P4-Lev	40	0.543	0.743	1.052
9		60	0.476	0.661	1.153
10		80	0.453	0.633	1.253
11		10	0.587	0.797	0.850
12		20	0.554	0.757	0.964
13	P6-Lev	40	0.509	0.702	1.090
14		60	0.464	0.647	1.116
15		80	0.457	0.675	1.140
16		10	0.652	0.876	0.699
17		20	0.609	0.824	0.800
18	PA2-Lev	40	0.554	0.757	0.913
19		60	0.509	0.702	1.014
20		80	0.498	0.688	1.128
21		10	0.663	0.889	0.713
22		20	0.620	0.837	0.839
23	PA4-Lev	40	0.521	0.716	1.027
24		60	0.509	0.702	1.090
25		80	0.498	0.688	1.152
26		10	0.587	0.797	0.850
27	PA6-Lev	20	0.554	0.757	0.939
28		40	0.521	0.716	1.027

29	60	0.487	0.675	1.090
30	80	0.464	0.647	1.166

[a]The Kamlet-Taft empirical parameters was the average of twice measurements, and the values in the parentheses were the half of the range of replicate results

Table S11. The Kamlet-Taft empirical parameters of the DMI/PDESs.[a]

Entry	PDES	DMI content (vol%)	π*	α	β
1		10	0.673	0.902	0.727
2		20	0.630	0.850	0.775
3	P2-Lev	40	0.565	0.770	0.823
4		60	0.521	0.716	0.899
5		80	0.441	0.619	0.935
6		10	0.609	0.824	1.028
7		20	0.587	0.797	0.978
8	P4-Lev	40	0.554	0.757	0.913
9		60	0.521	0.716	0.925
10		80	0.453	0.633	0.923
11		10	0.554	0.757	0.964
12		20	0.532	0.73	0.938
13	P6-Lev	40	0.509	0.70	0.911
14		60	0.464	0.65	0.936
15		80	0.418	0.59	0.961
16		10	0.673	0.902	0.568
17		20	0.652	0.876	0.565
18	PA2-Lev	40	0.630	0.850	0.535
19		60	0.543	0.743	0.715
20		80	0.441	0.619	0.909
21		10	0.630	0.850	0.697
22		20	0.587	0.797	0.745
23	PA4-Lev	40	0.532	0.729	0.834
24		60	0.476	0.661	0.871
25		80	0.418	0.591	0.935
26		10	0.576	0.784	0.836
27		20	0.532	0.729	0.860
28	PA6-Lev	40	0.487	0.675	0.884
29		60	0.441	0.619	0.935
30		80	0.407	0.577	0.974

[a]The Kamlet-Taft empirical parameters was the average of twice measurements, and the values in the parentheses were the half of the range of replicate results

Table S12. The Kamlet-Taft empirical parameters of the Cyrene/PDESs.a

Entry	PDES	Cyrene content (vol%)	π*	α	β
1		10	0.727	0.967	0.771
2		20	0.694	0.928	0.755
3	P2-Lev	40	0.663	0.889	0.713
4		60	0.620	0.837	0.761
5		80	0.587	0.797	0.824
6		10	0.673	0.902	0.804
7		20	0.652	0.876	0.803
8	P4-Lev	40	0.630	0.850	0.827
9		60	0.620	0.837	0.813
10		80	0.598	0.810	0.864
11		10	0.587	0.797	1.003
12		20	0.586	0.784	0.990
13	P6-Lev	40	0.577	0.797	0.952
14		60	0.567	0.797	0.927
15		80	0.556	0.784	0.888
16		10	0.908	1.190	0.384
17		20	0.878	1.141	0.429
18	PA2-Lev	40	0.778	1.092	0.474
19		60	0.673	1.055	0.508
20		80	0.576	0.837	0.735
21		10	0.888	1.166	0.537
22		20	0.878	1.153	0.548
23	PA4-Lev	40	0.778	1.030	0.661
24		60	0.673	0.902	0.779
25		80	0.576	0.784	0.836
26		10	0.576	0.784	0.939
27		20	0.576	0.784	0.914
28	PA6-Lev	40	0.576	0.784	0.888
29		60	0.565	0.770	0.875
30		80	0.576	0.784	0.836

[a]The Kamlet-Taft empirical parameters was the average of twice measurements, and the values in the parentheses were the half of the range of replicate results

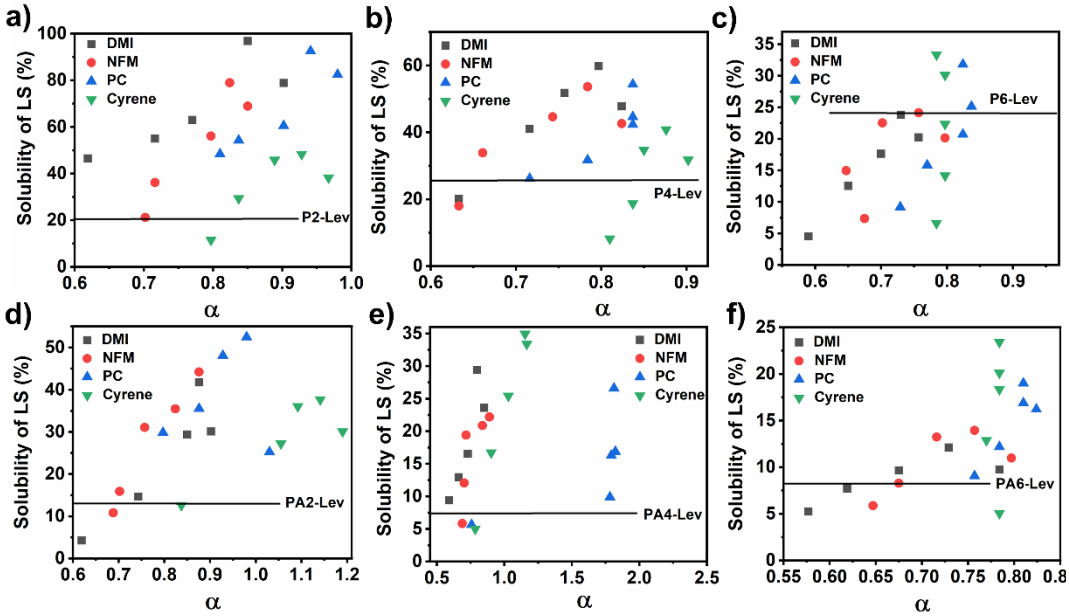

Fig. S4. The correlation between the LS solubility and the α value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents).

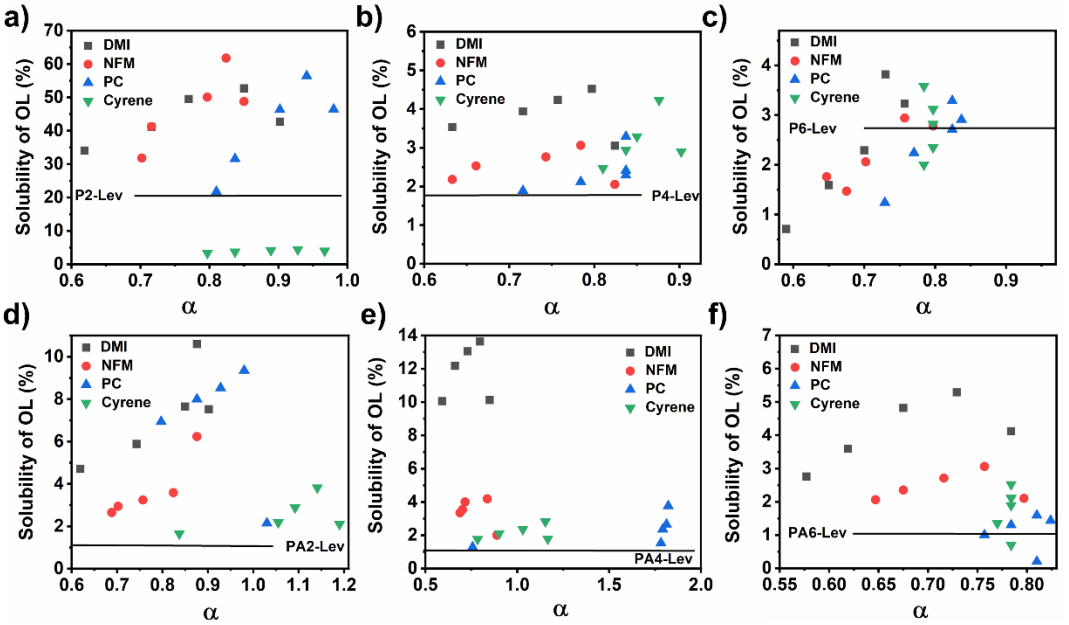

Fig. S5. The correlation between the OL solubility and the α value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents).

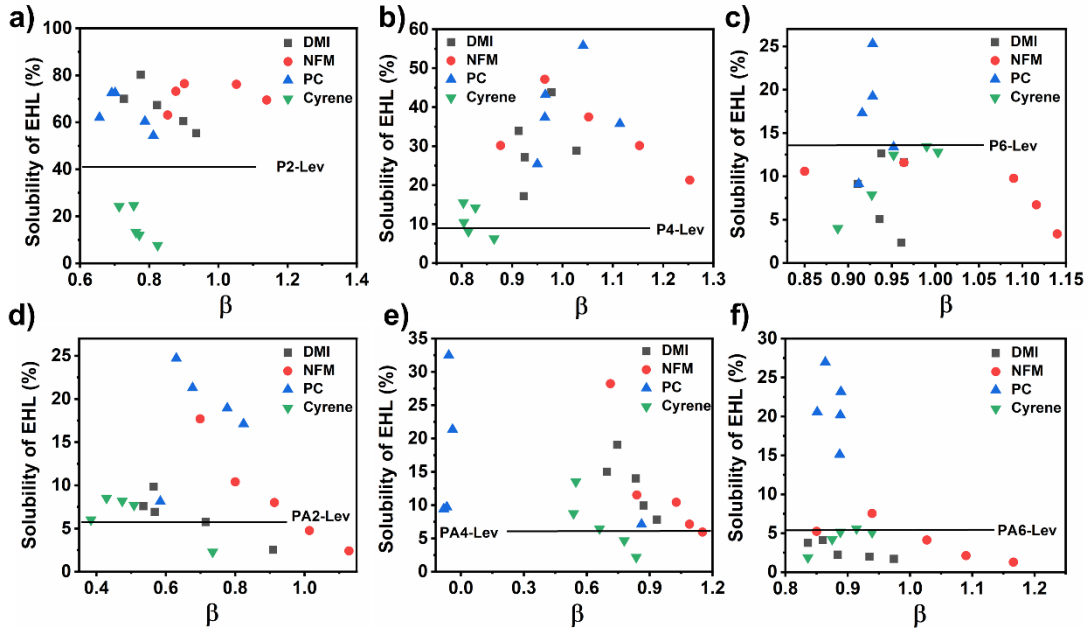

Fig. S6. The correlation between the EHL solubility and the β value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents).

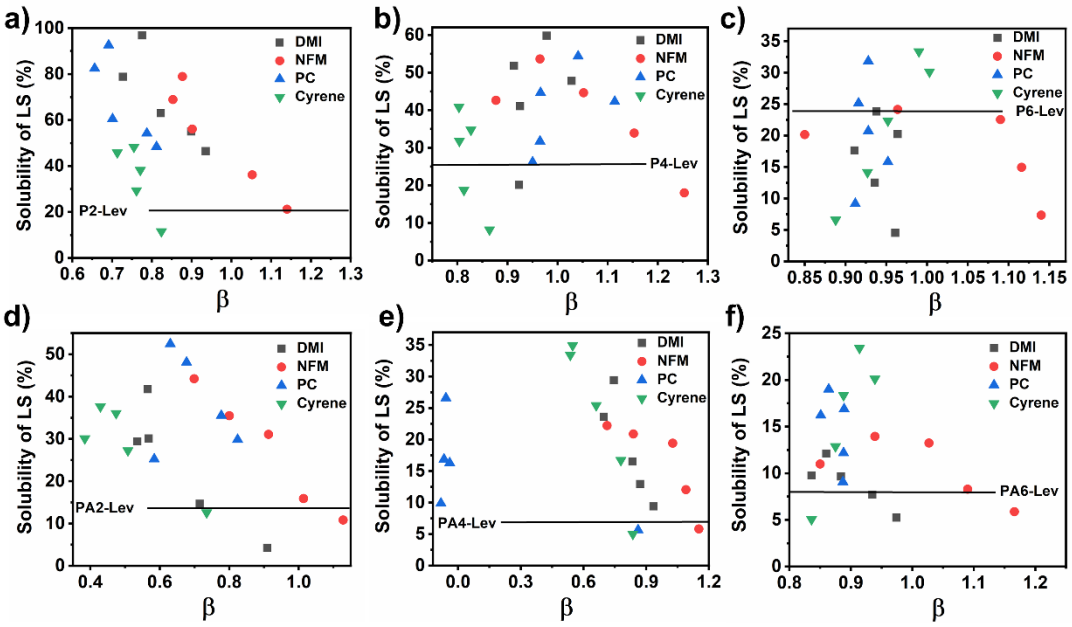

Fig. S7. The correlation between the LS solubility and the β value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents).

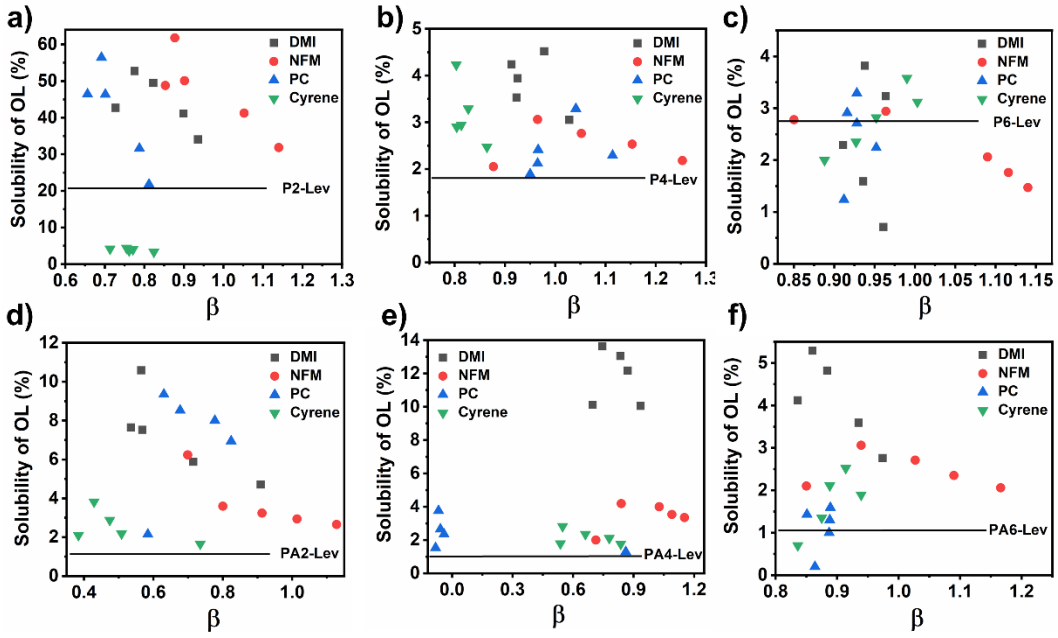

Fig. S8. The correlation between the OL solubility and the β value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents).

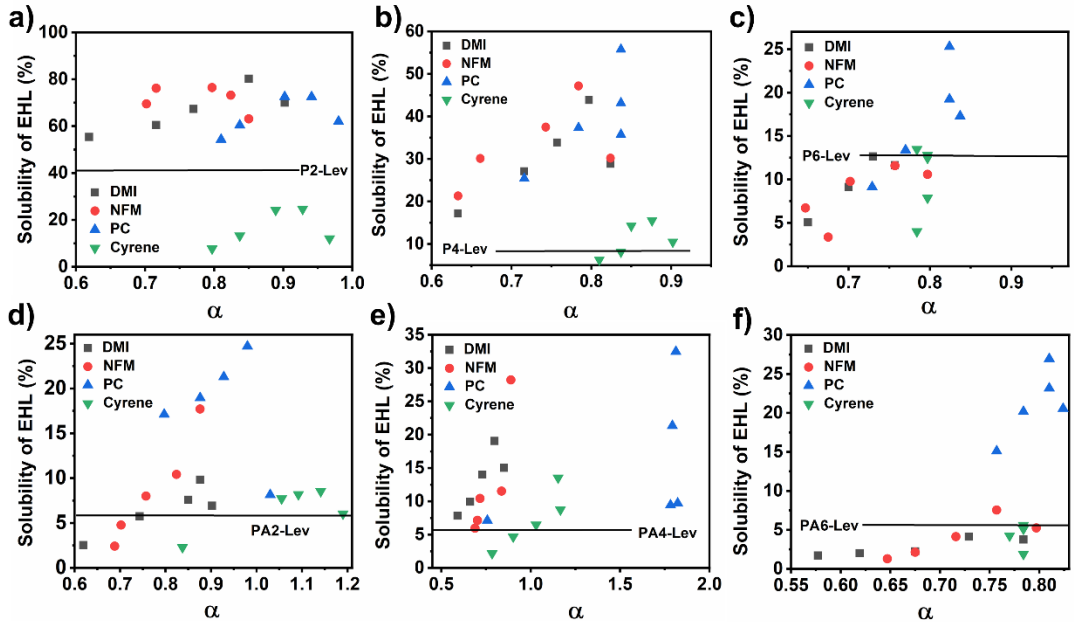

Fig. S9. The correlation between the EHL solubility and the α value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents).

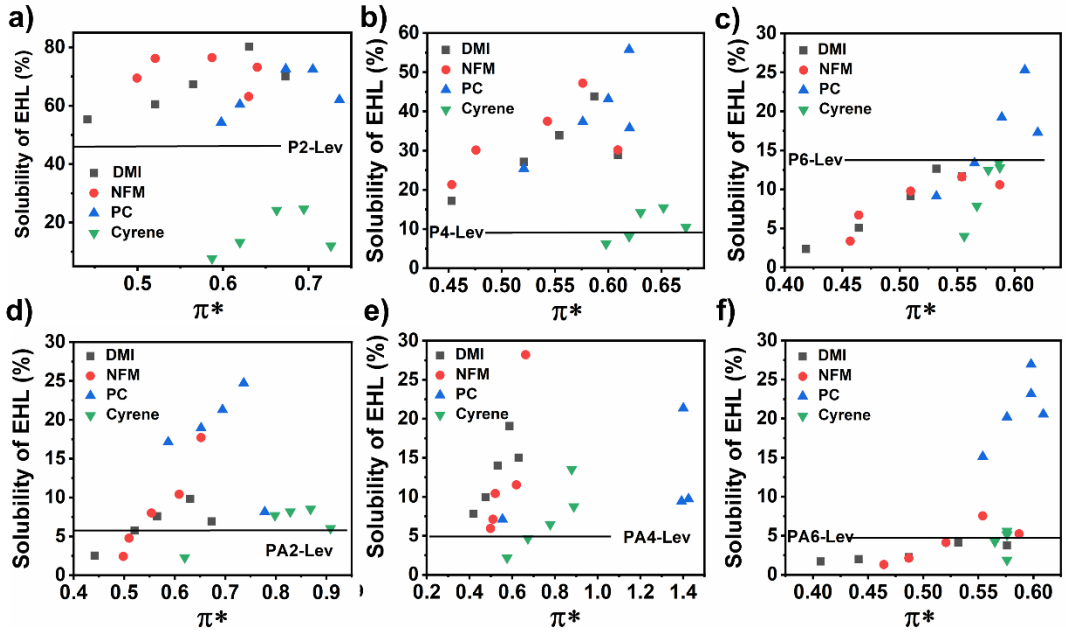

Fig. S10. The correlation between the EHL solubility and the π* value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents, and the critical value of π* was 0.441, 0.452, 0.587, 0.520, 0.418 and 0.554 in P2-Lev, P4-Lev, P6-Lev, PA2-Lev, PA4-Lev, and PA6-Lev, respectively).

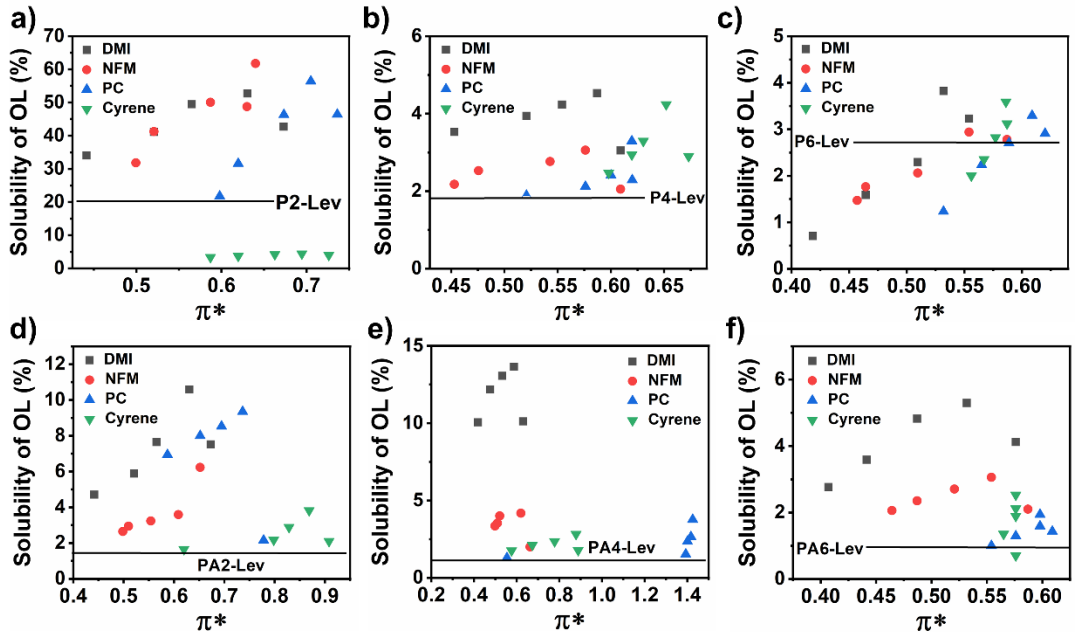

Fig. S11. The correlation between the OL solubility and the π* value of the corresponding solvent (the line represents the solubility of the lignin in PDESs, and the data above the line indicated that lignin was promoted to dissolve by adding co-solvents, and the critical value of π* was 0.441, 0.452, 0.531, 0.441, 0.418 and 0.406 in P2-Lev, P4-Lev, P6-Lev, PA2-Lev, PA4-Lev, and PA6-Lev, respectively).

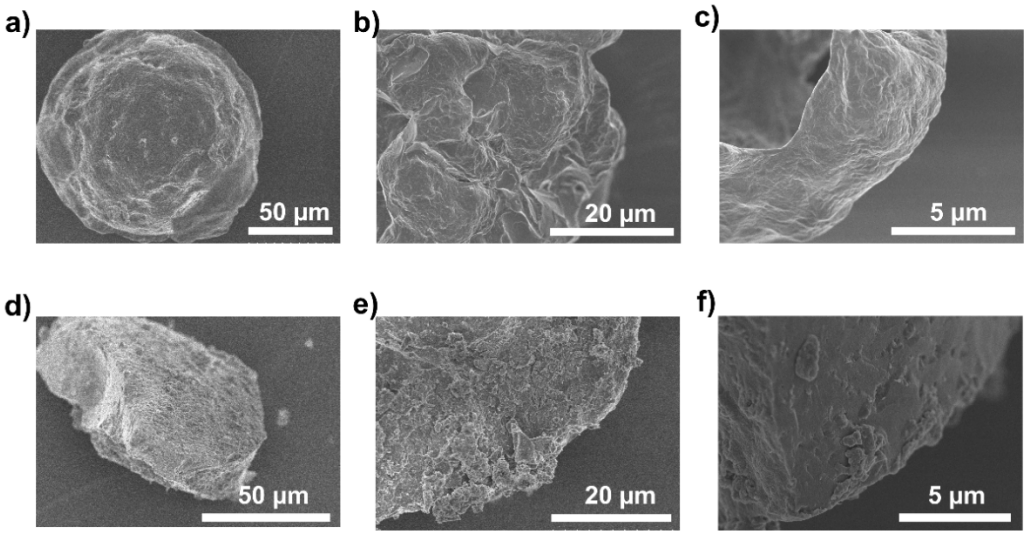

Fig. S12. SEM images of a), b) and c) initial LS and d), e) and f) regenerated LS.

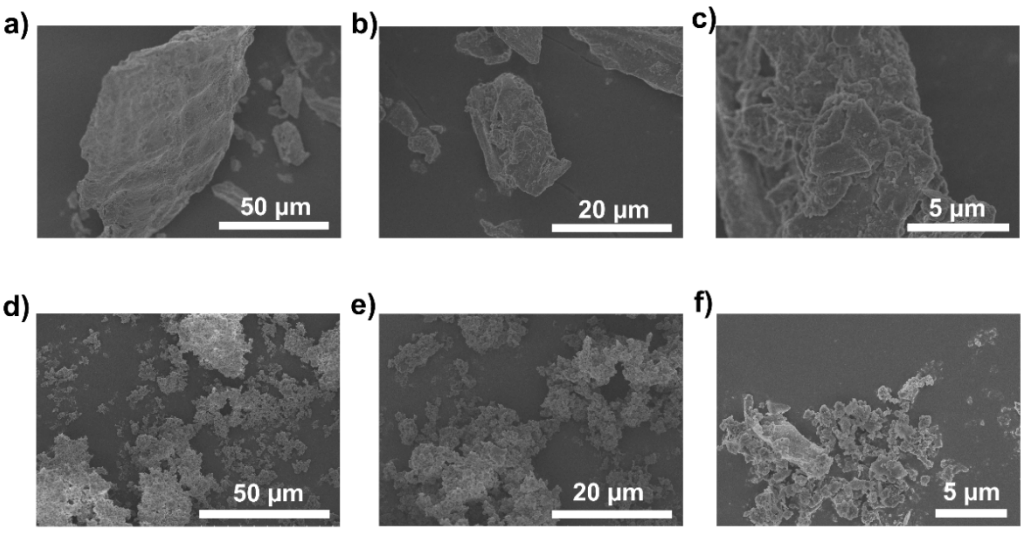

Fig. S13. SEM images of a), b) and c) initial OL and d), e) and f) regenerated OL.

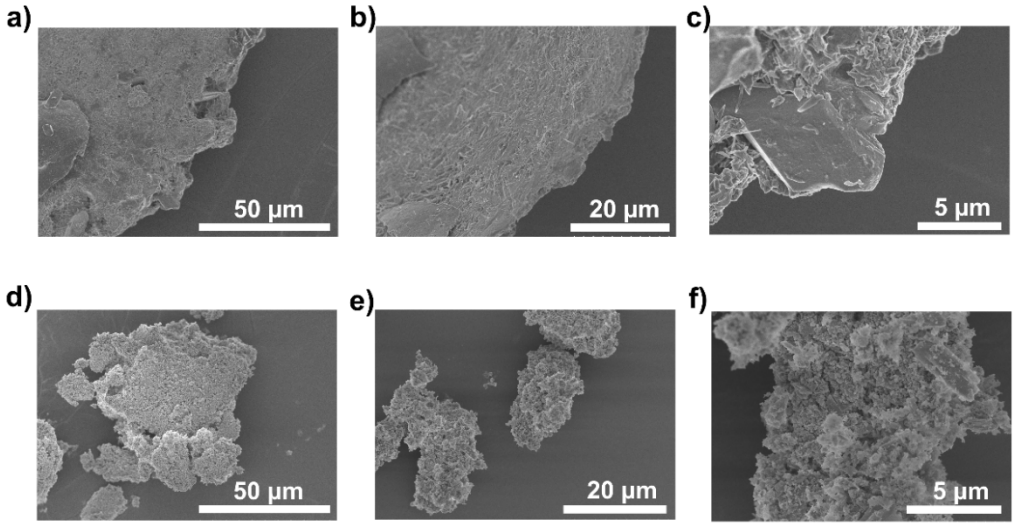

Fig. S14. SEM images of a), b) and c) initial EHL and d), e) and f) regenerated EHL.

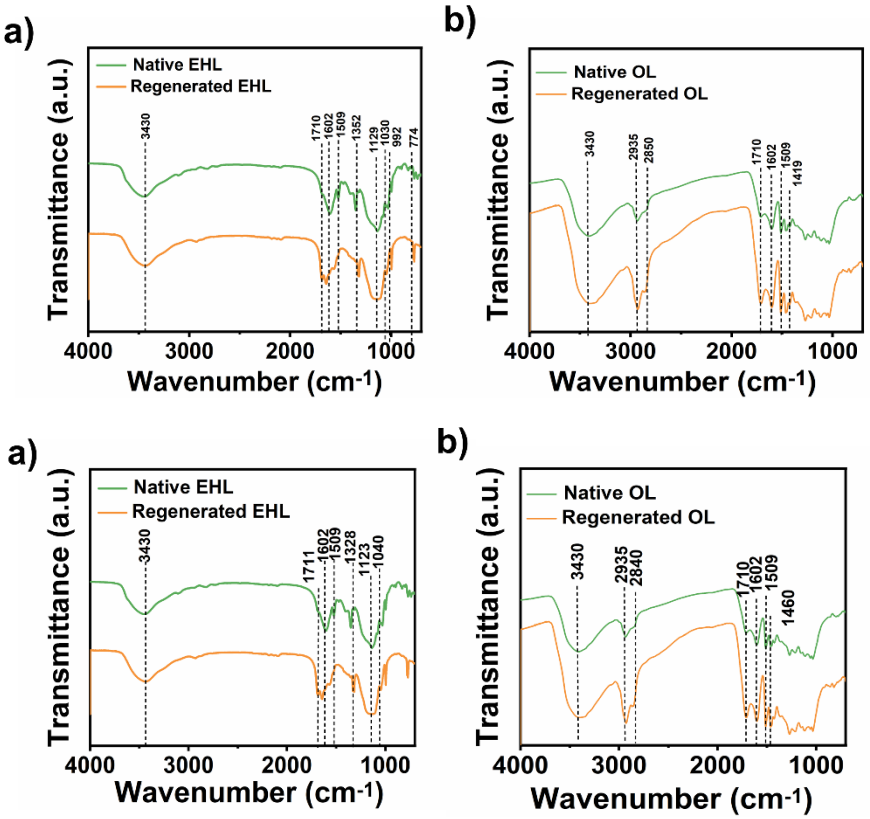

Fig. S15. The FT-IR of native and generated lignins

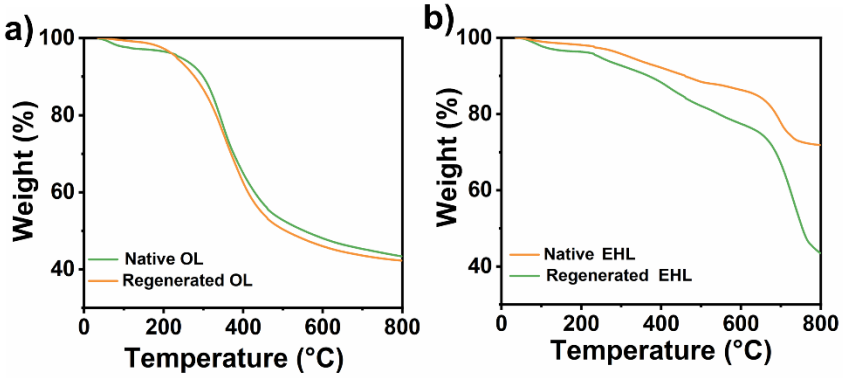

Fig. S16. TGA of initial and regenerated lignin

Table S13. Sugar content and molecular weight of three lignin samples.

Samples	Sugar (%)						Purity (%)	Molecular weight (g mol^{-1})		PDI[a]
	Glucose	Xylose	Arabinose	Galactose	Uronic acid	Total sugars		M_w	M_n	
EHL	2.27	ND[b]	ND	ND	0.16	2.43	97.57	2173	1145	1.90
OL	ND	0.28	0.06	ND	0.02	0.36	99.64	2521	1240	2.03
LS	0.32	2.53	1.95	1.04	0.05	5.89	94.11	1413	1258	1.12

[a] PDI: Polydispersity index.

[b] ND: Not Detected.

Table S14. Element analysis of different lignin

Sample	Mass fraction (%)				
	N	**C**	**H**	**S**	**O**
EHL	0.67	59.27	5.74	0.16	20.00
LS	0.66	31.26	3.97	6.40	24.62
OL	0.38	62.00	6.19	0.04	20.30