Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Multiscale Solvation Effect on Reactivity of β-O-4 of Lignin Dimers in Deep Eutectic Solvents

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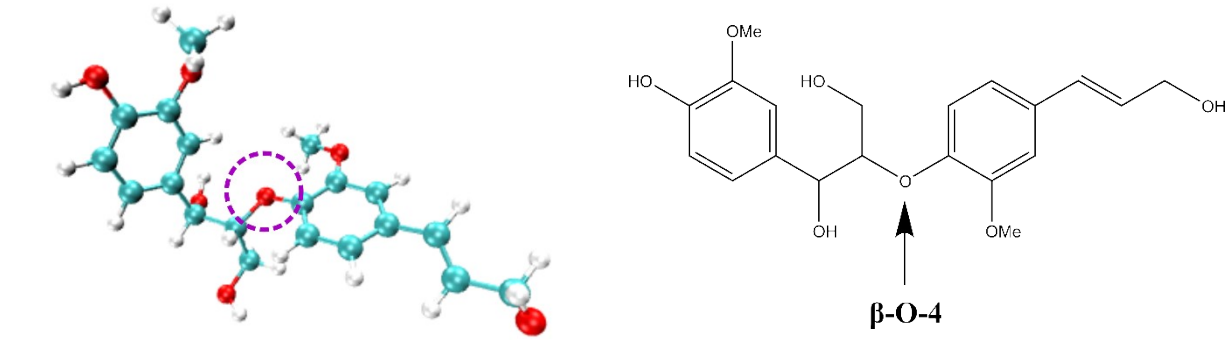
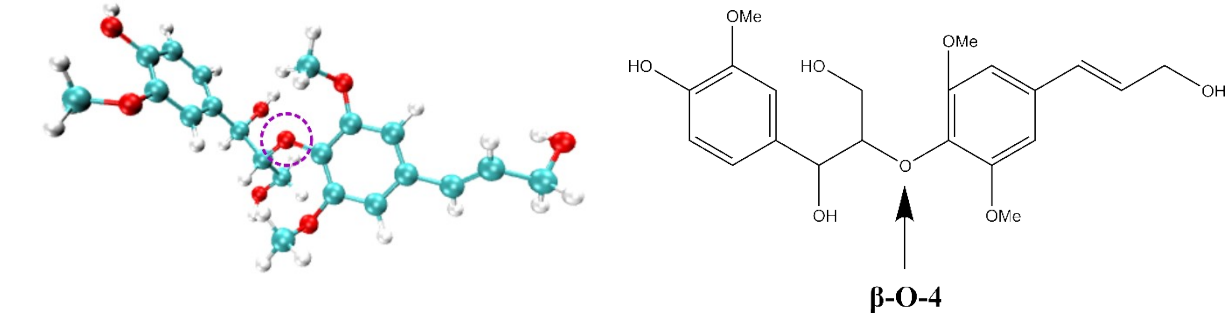


(a)



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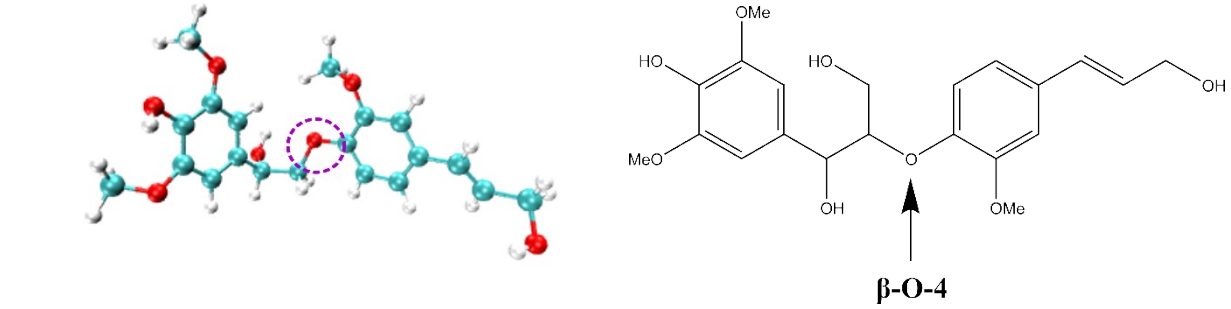
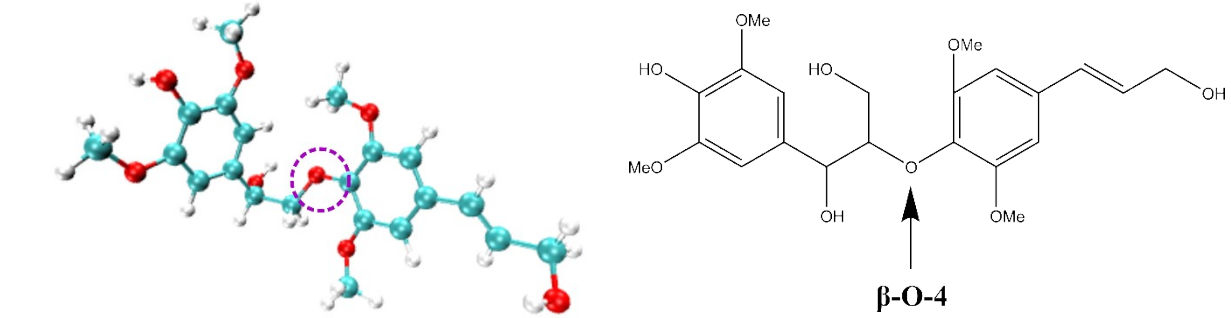


(c)



(d)

Figure S1. The configuration and chemical sketch of (a) G-G, (b) G-S, (c) S-G and (d) S-S lignin dimer.

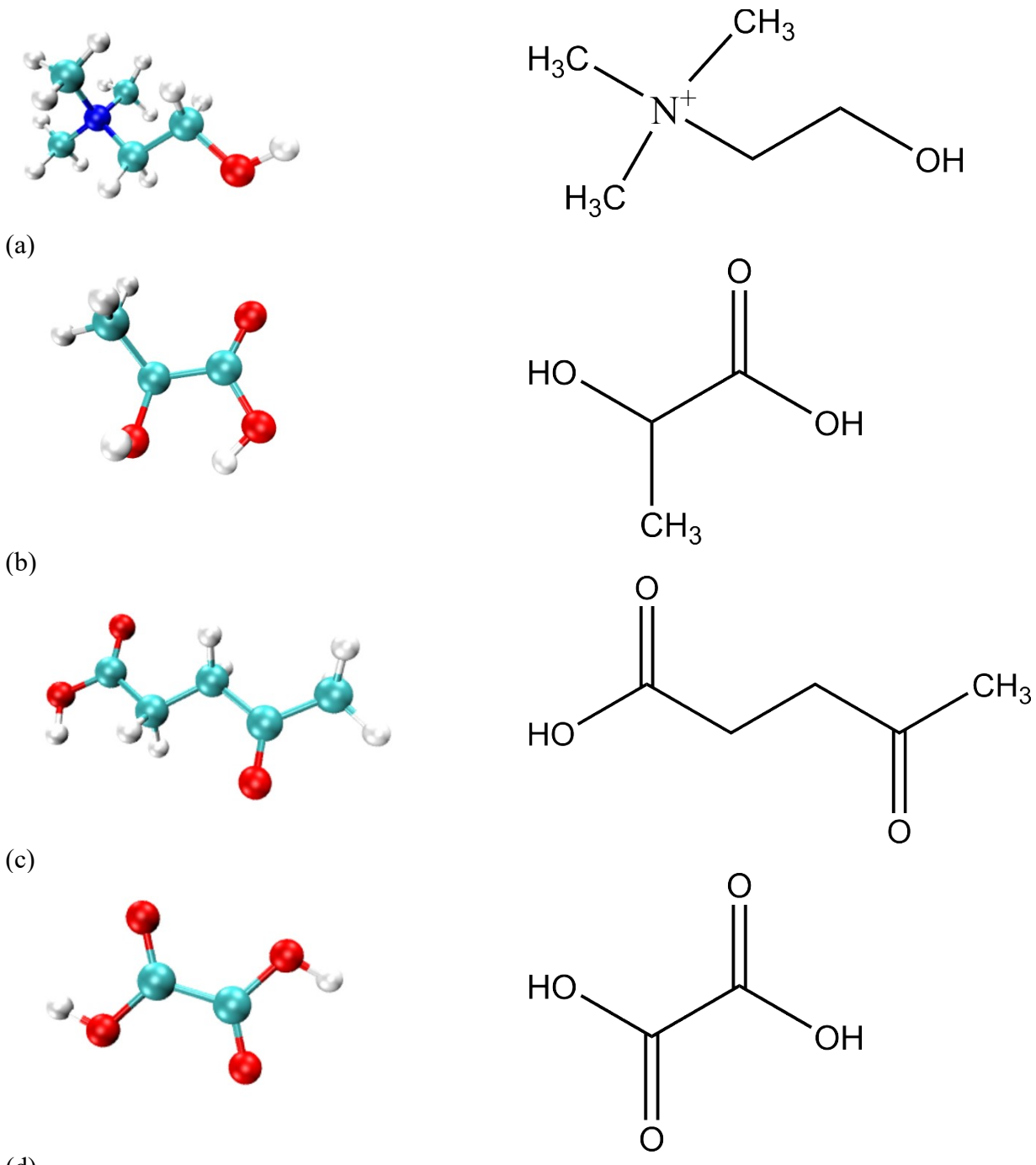
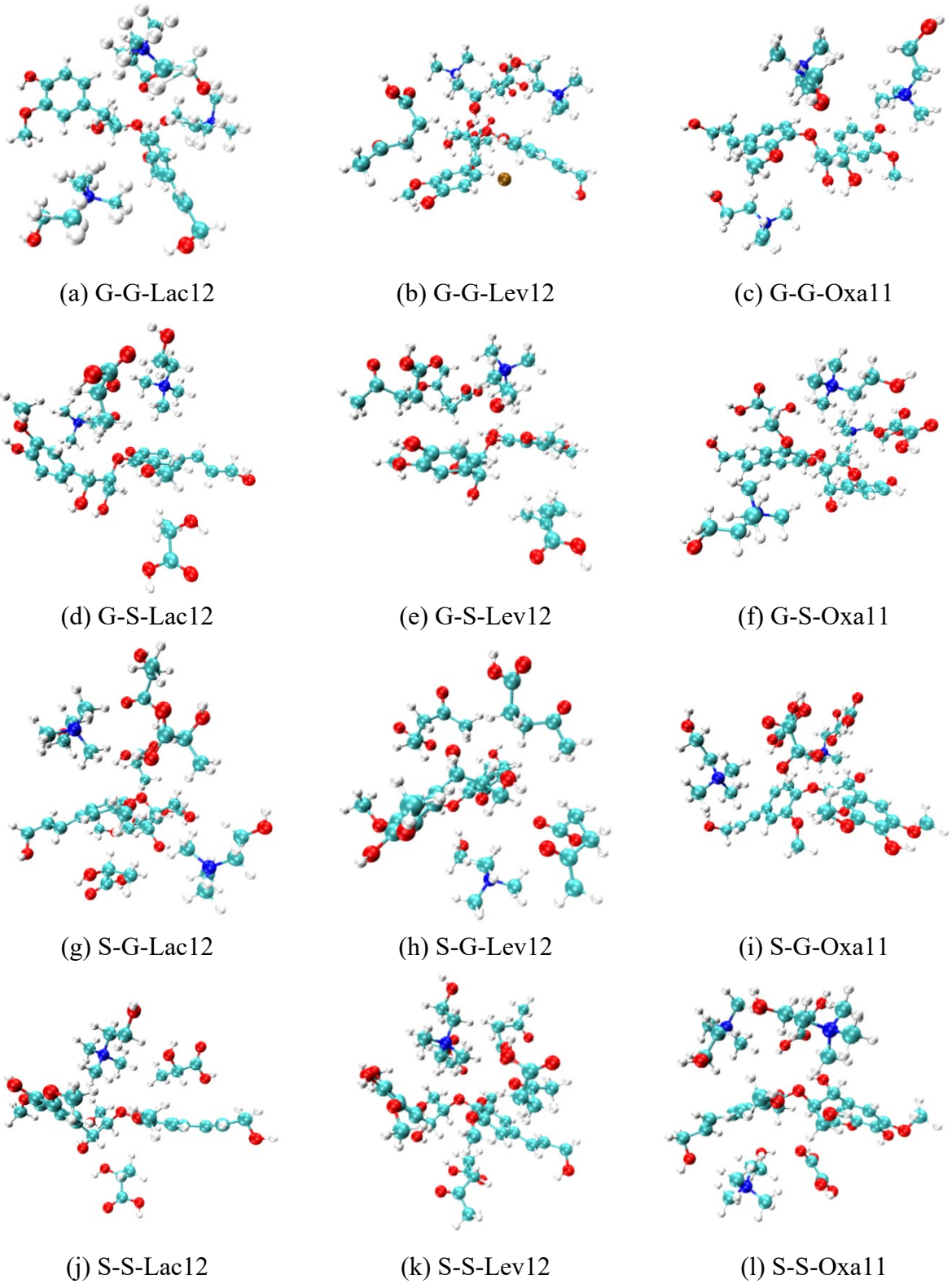


Figure S2. The configuration and chemical sketch of (a) choline, (b) lactic acid, (c) levulinic acid and (d) oxalic acid.



(a) G-G-Lac12 (b) G-G-Lev12 (c) G-G-Oxa11

(d) G-S-Lac12 (e) G-S-Lev12 (f) G-S-Oxa11

(g) S-G-Lac12 (h) S-G-Lev12 (i) S-G-Oxa11

(j) S-S-Lac12 (k) S-S-Lev12 (l) S-S-Oxa11

Figure S3. the snapshots of the configuration used in DFT in which the β-O_4 present the largest SASA for the twelve systems. Color representations are the same as in Figure 1.

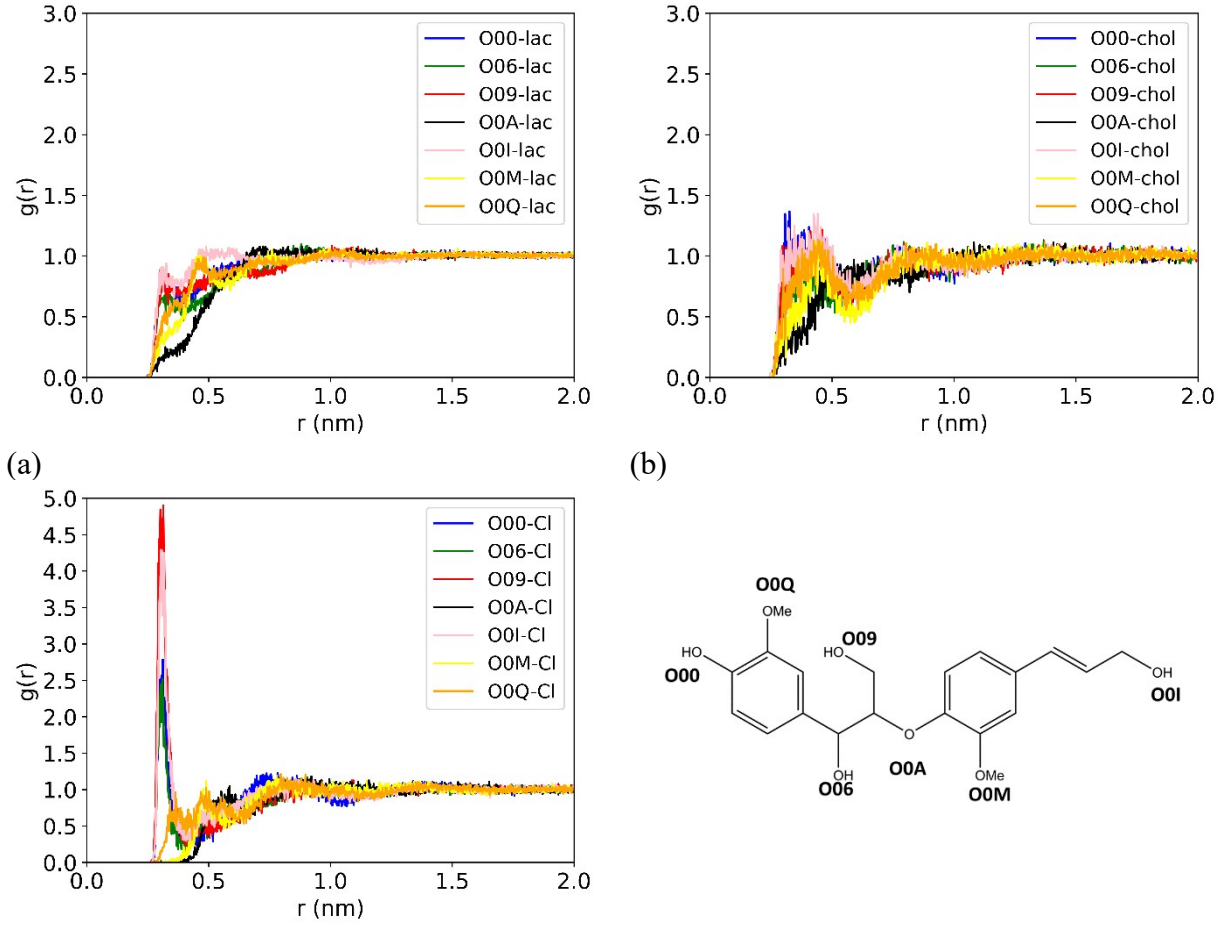


Figure S4. The radial distribution function (RDF) of seven oxygen atoms of G-G lignin dimer and (a) all oxygen atoms on lactic acid, (b) oxygen atom of choline and (c) Cl^-. The seven oxygen atoms of G-G dimer used in RDF are labeled in (d).

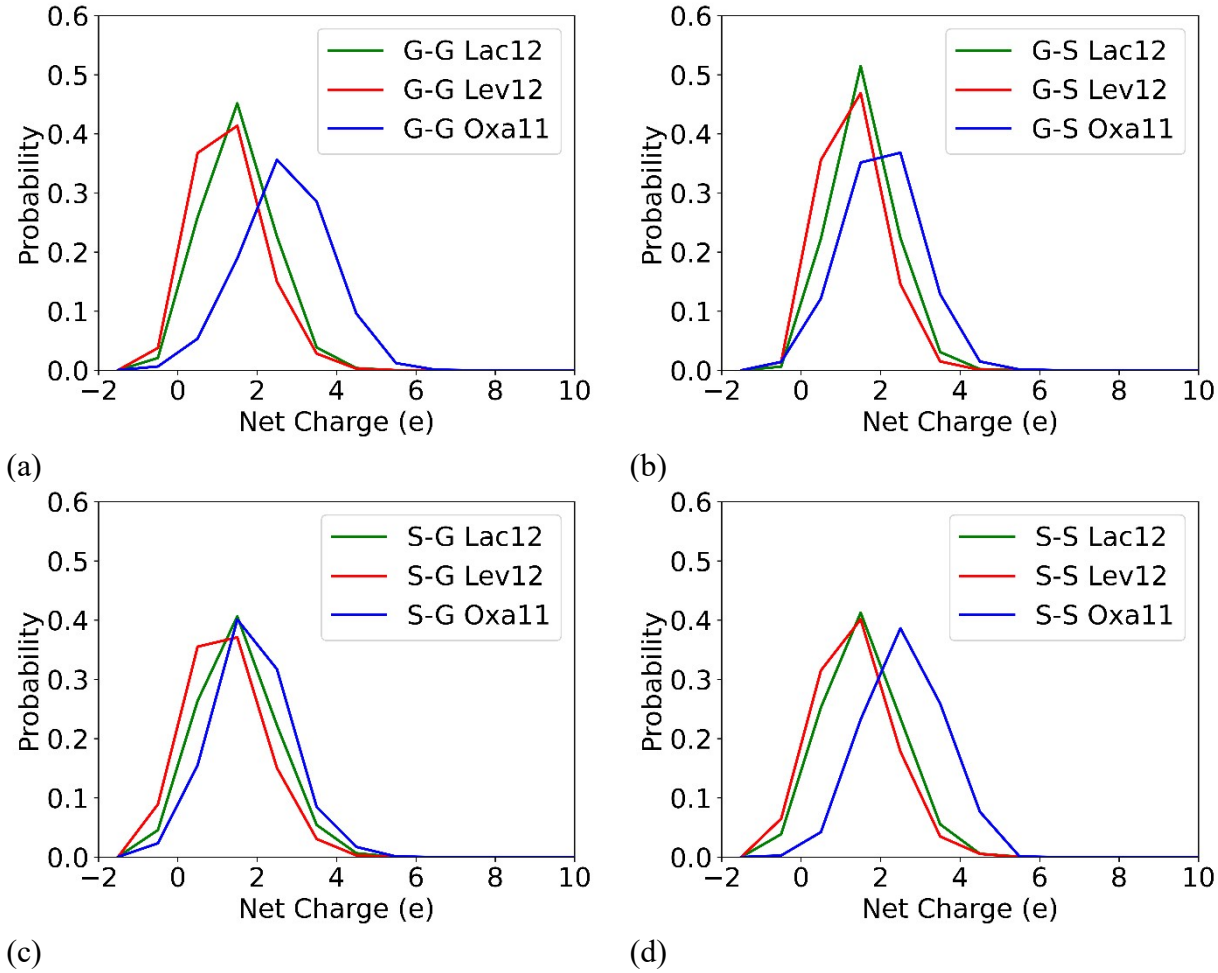


Figure S5. The distribution of net charge of the solvation shell around the β-O-4 linkage of (a) G-G, (b) G-S, (c) S-G and (d) S-S lignin dimers in three DESs.

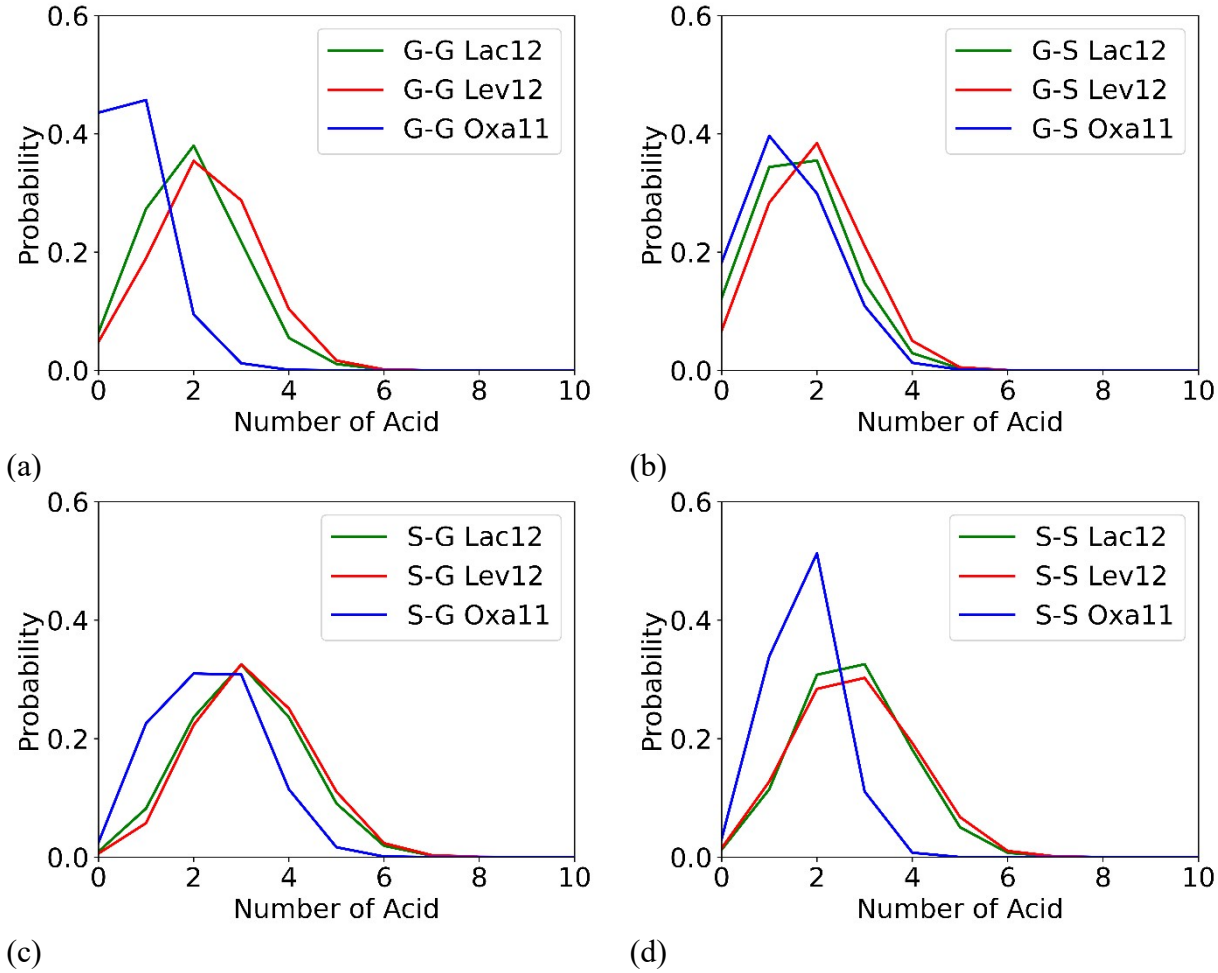


Figure S6. The distribution of acid molecules of the solvation shell around the β-O-4 linkage of (a) G-G, (b) G-S, (c) S-G and (d) S-S lignin dimers in three DESs.

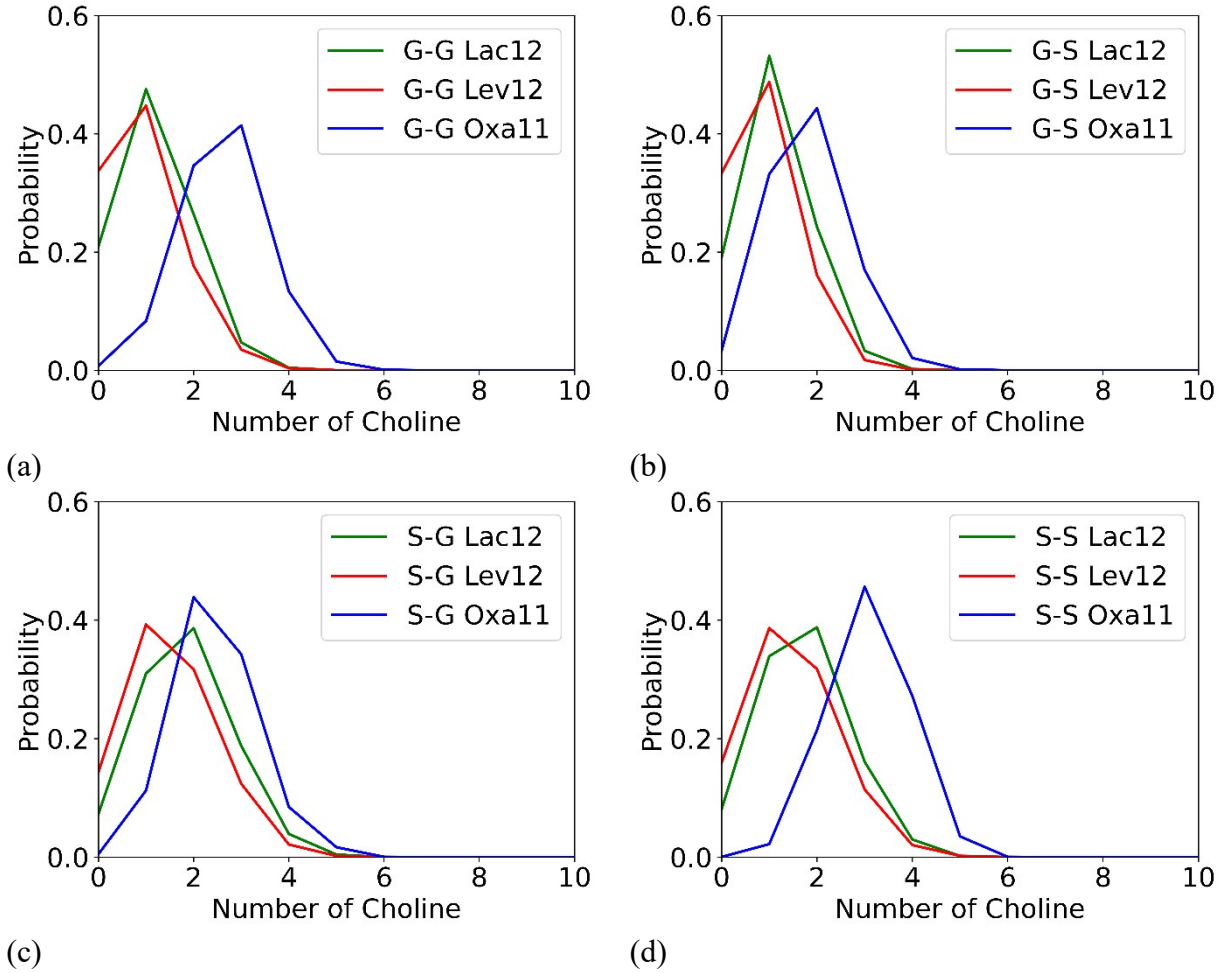


Figure S7. The distribution of choline molecules of the solvation shell around the β-O-4 linkage of (a) G-G, (b) G-S, (c) S-G and (d) S-S lignin dimers in three DESs.

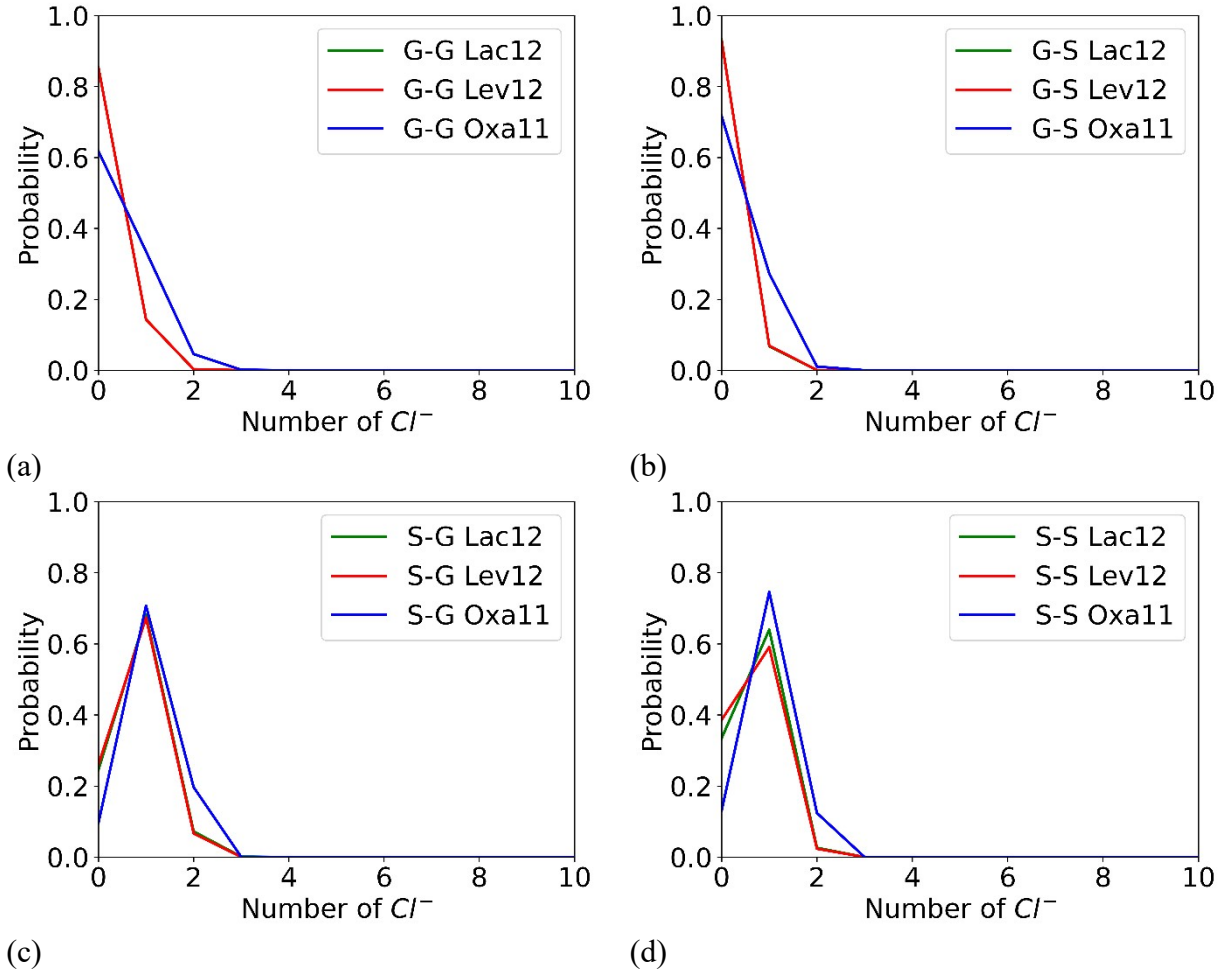
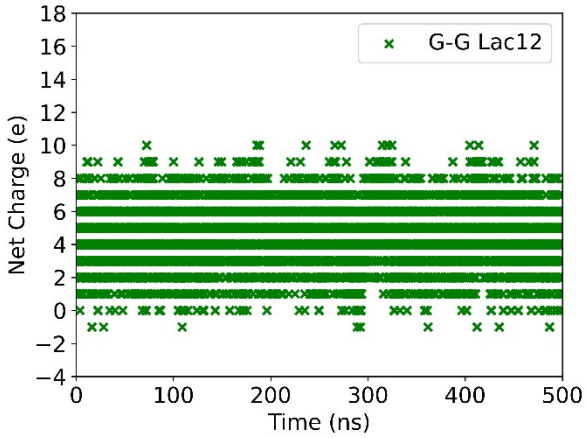
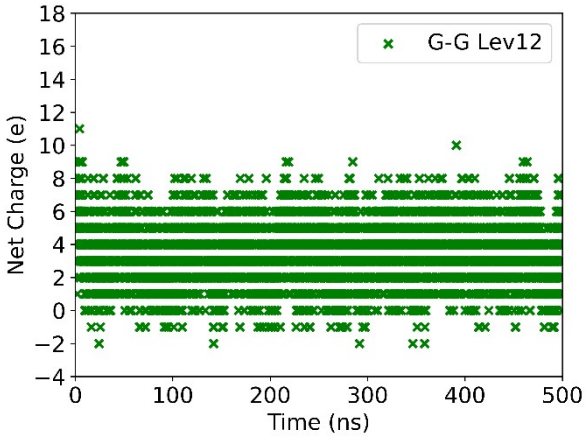


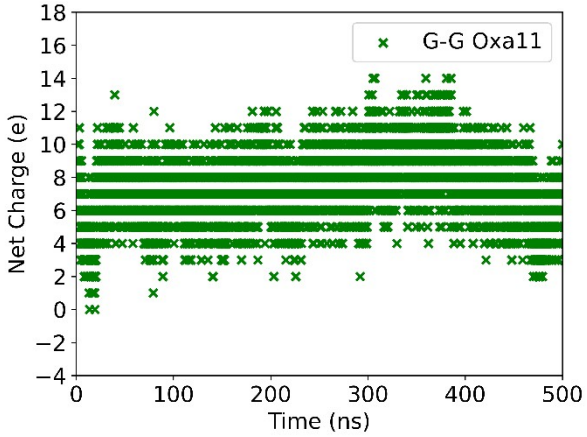
Figure S8. The distribution of Cl^- ions of the solvation shell around the β-O-4 linkage of (a) G-G, (b) G-S, (c) S-G and (d) S-S lignin dimers in three DESs.



(a) G-G-Lac12

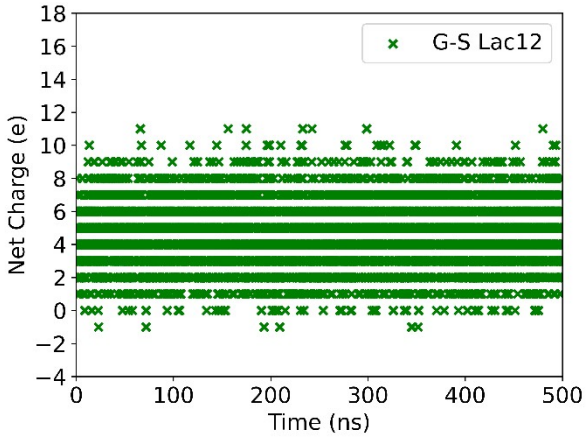


(b) G-G-Lev12

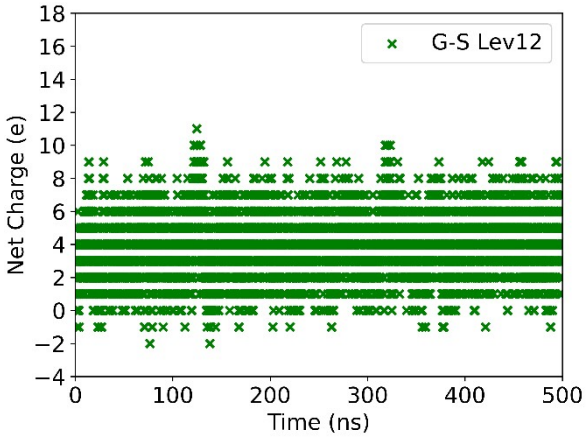


(c) G-G-Oxa11

Figure S9. The net charge of the solvation shell of G-G lignin dimer in the three DESs as a function of time.



(a) G-S-Lac12



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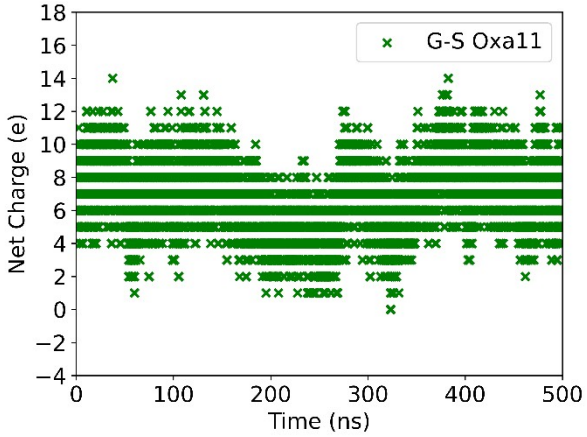
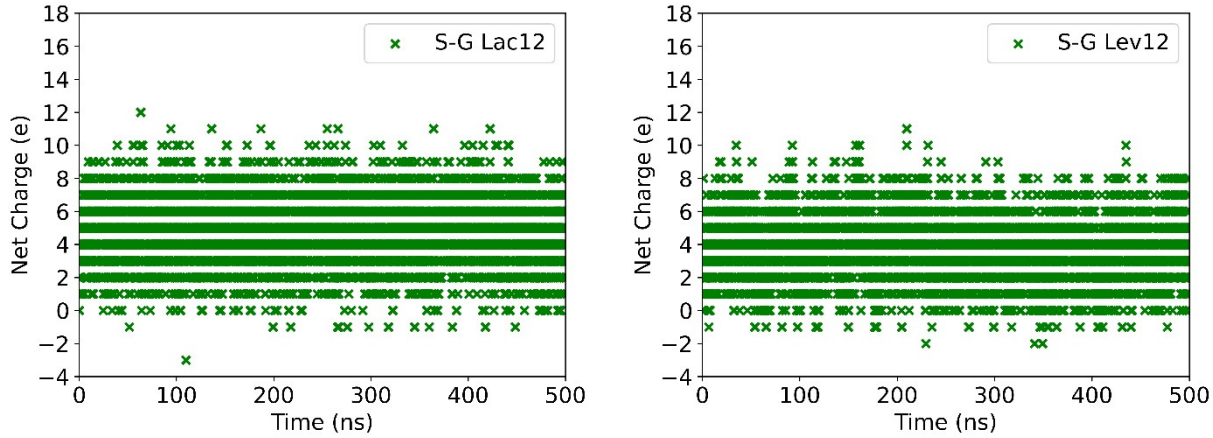


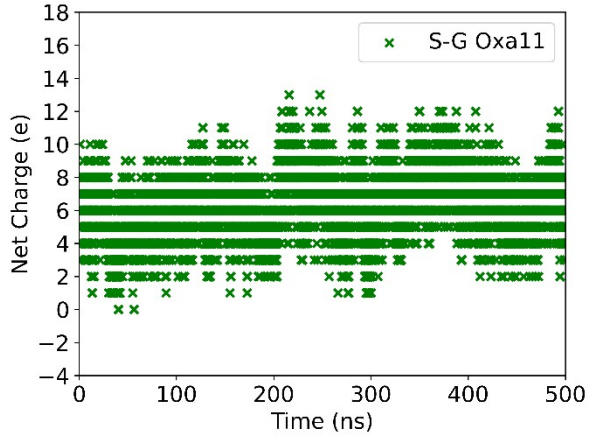
(c) G-S-Oxa11

Figure S10. The net charge of the solvation shell of G-S lignin dimer in the three DESs as a function of time.



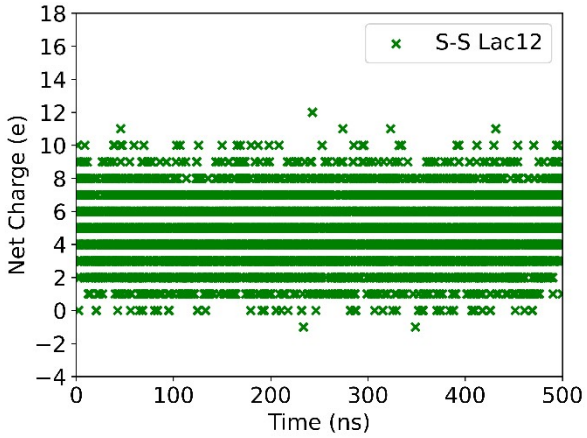
(a) S-G-Lac12

(b) S-G-Lev12

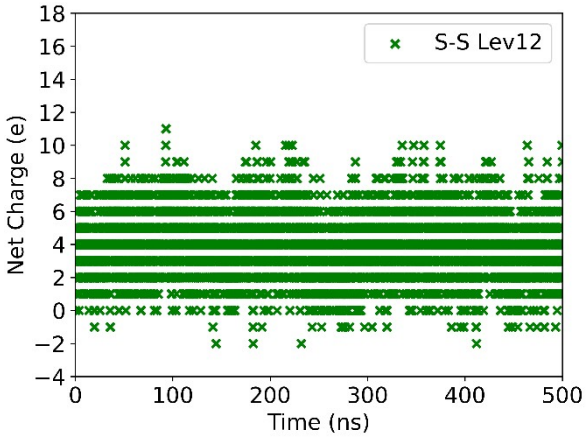


(c) S-G-Oxa11

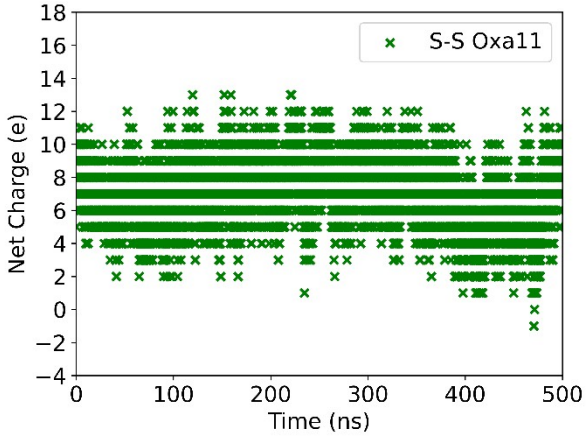
Figure S11. The net charge of the solvation shell of S-G lignin dimer in the three DESs as a function of time.



(a) S-S-Lac12



(b) S-S-Lev12



(c) S-S-Oxa11

Figure S12. The net charge of the solvation shell of S-S lignin dimer in the three DESs as a function of time.

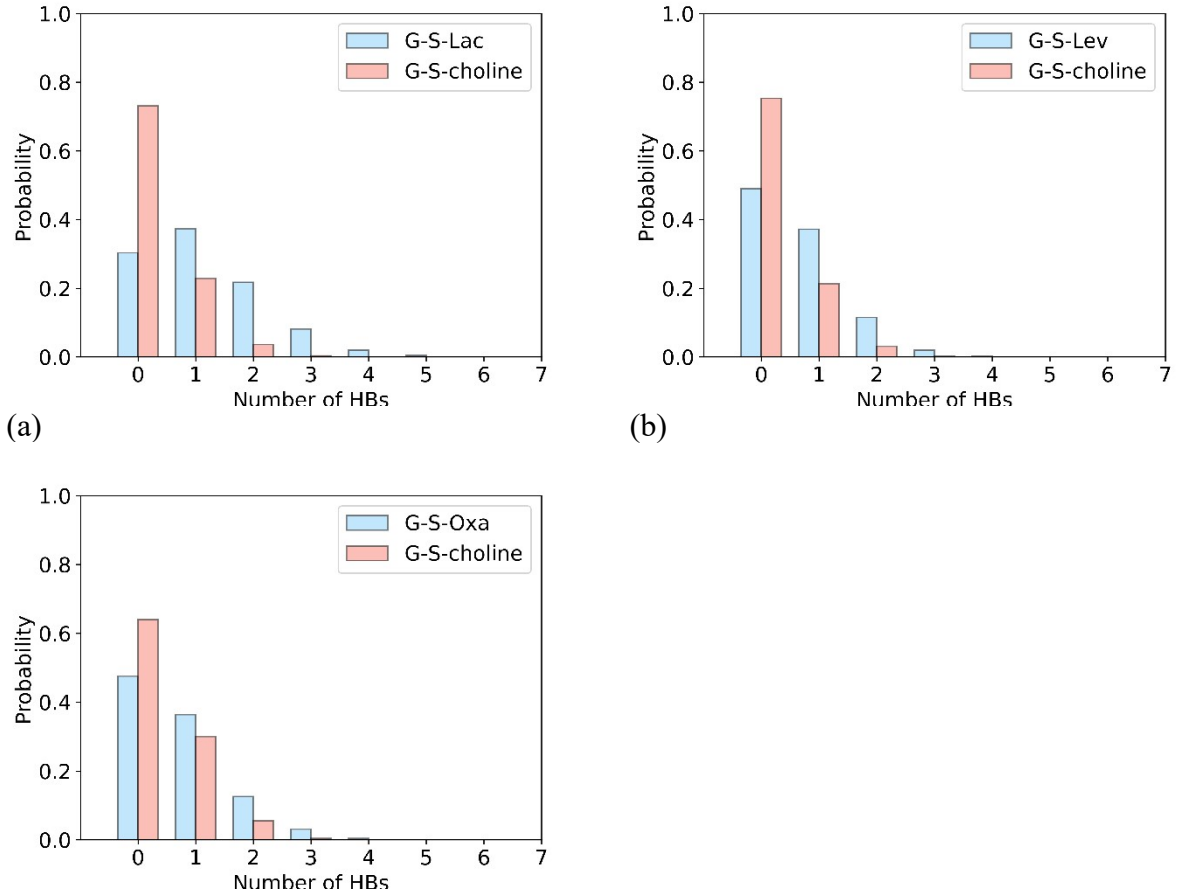


Figure S13. The distribution of HBs of lignin-acid and lignin-choline of (a) G-S Lac12, (b) G-S Lev12 and (c) G-S Oxa11.

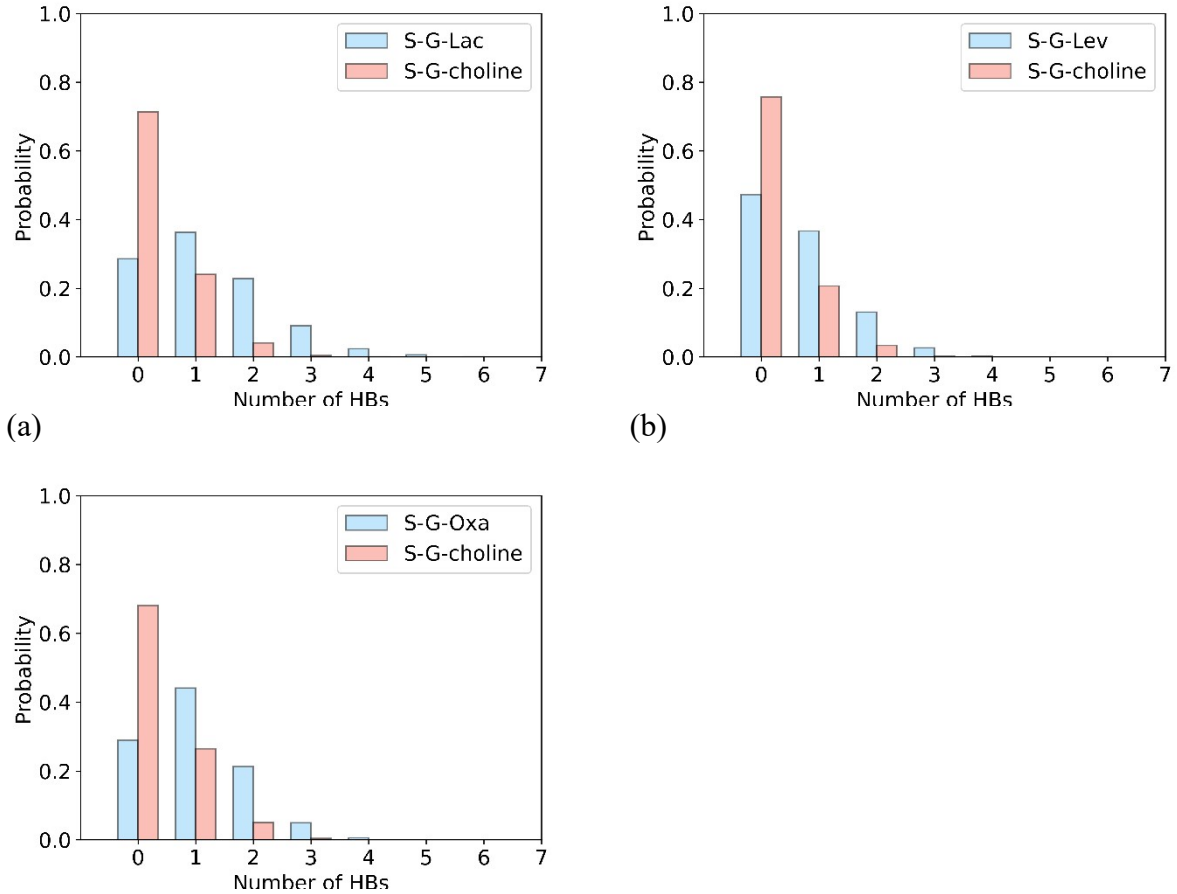


Figure S14. The distribution of HBs of lignin-acid and lignin-choline of (a) S-G Lac12, (b) S-G Lev12 and (c) S-G Oxa11.

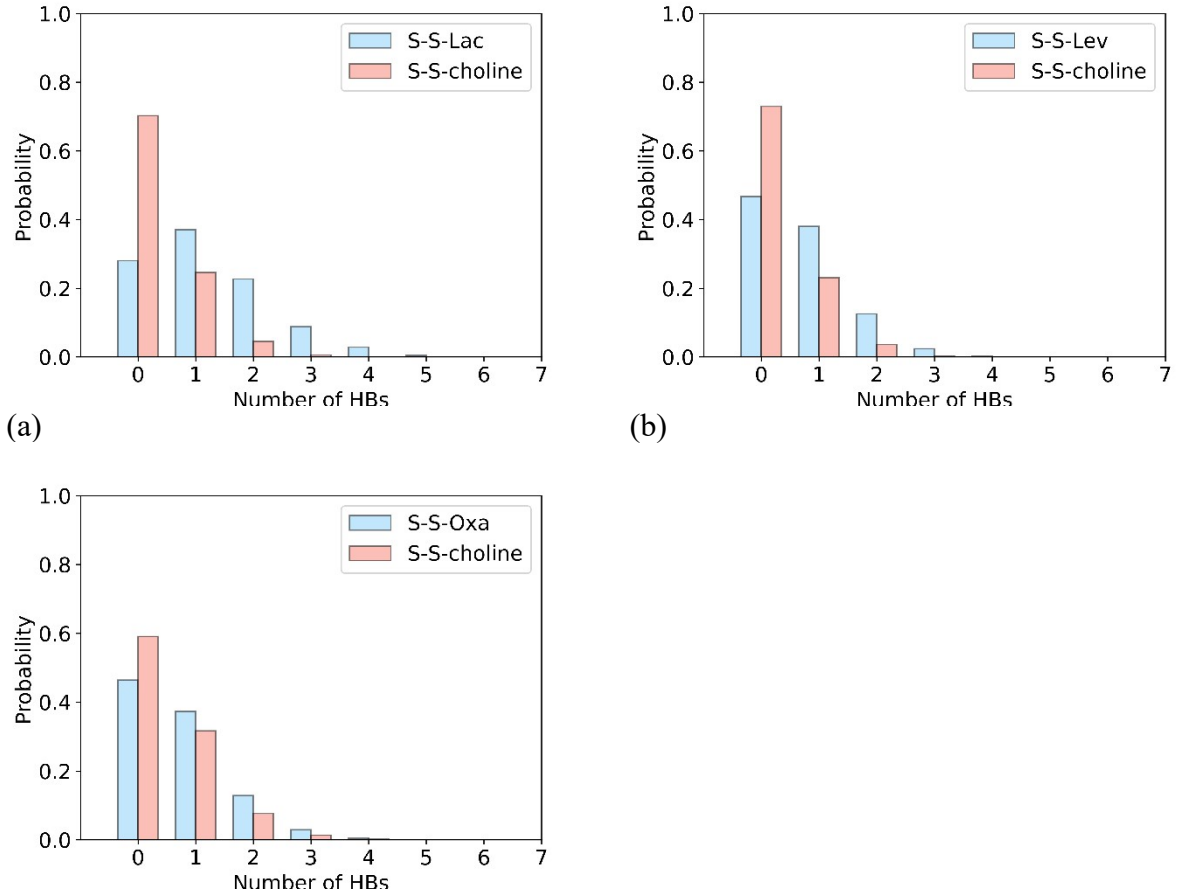


Figure S15. The distribution of HBs of lignin-acid and lignin-choline of (a) S-S Lac12, (b) S-S Lev12 and (c) S-S Oxa11.

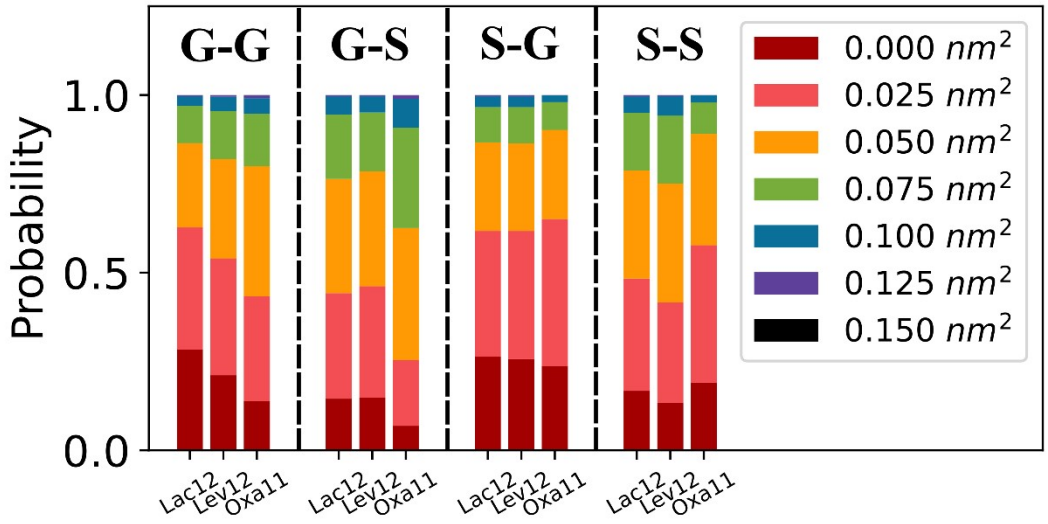
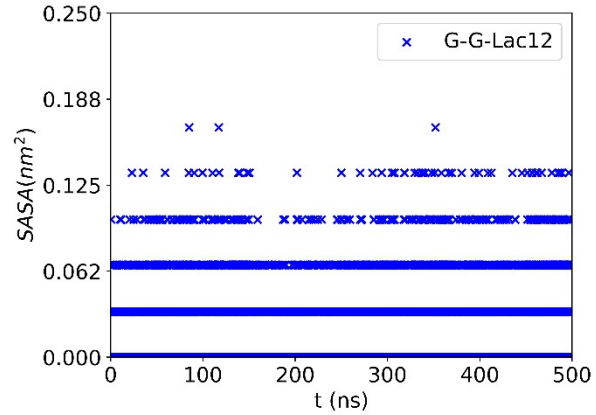
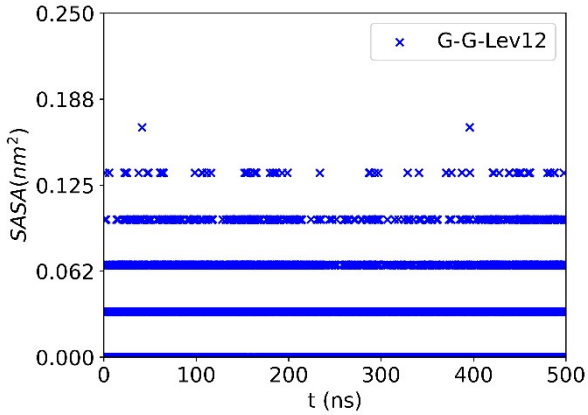


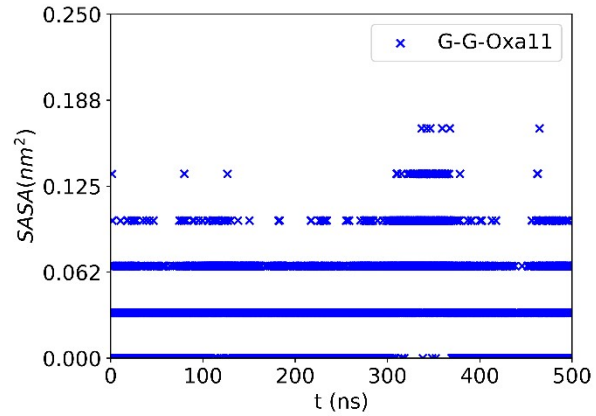
Figure S16. The distribution of SASA of β-O-4 for four types of lignin dimer in three DESs, the probe radius *r*=0.10 nm.



(a) G-G-Lac12

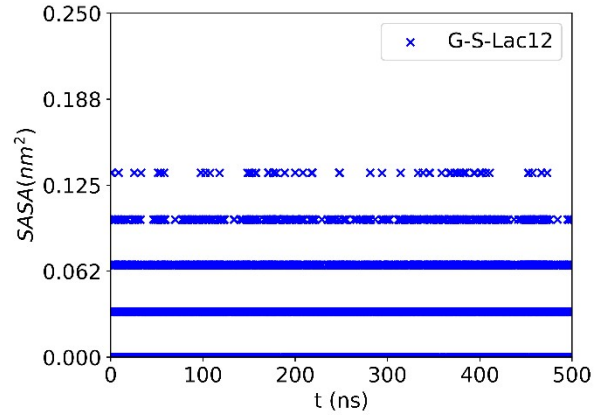


(b) G-G-Lev12

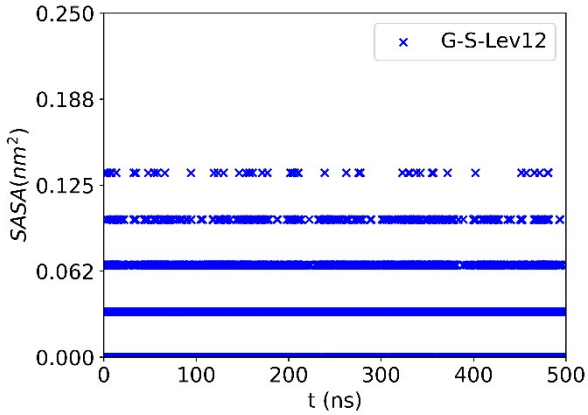


(c) G-G-Oxa11

Figure S17. The SASA of G-G lignin dimer in the three DESs as a function of time.



(a) G-S-Lac12



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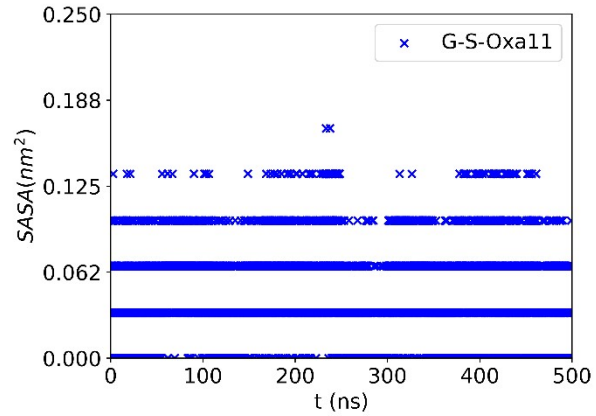
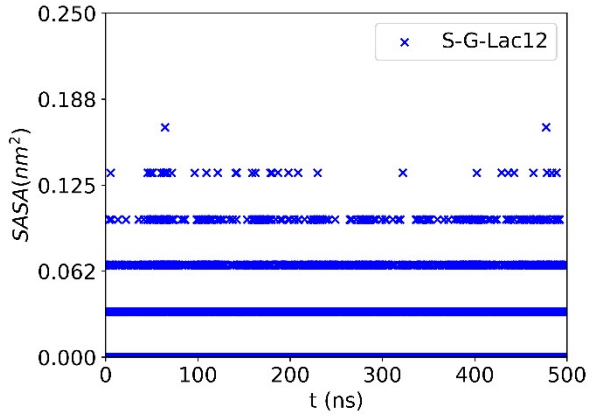
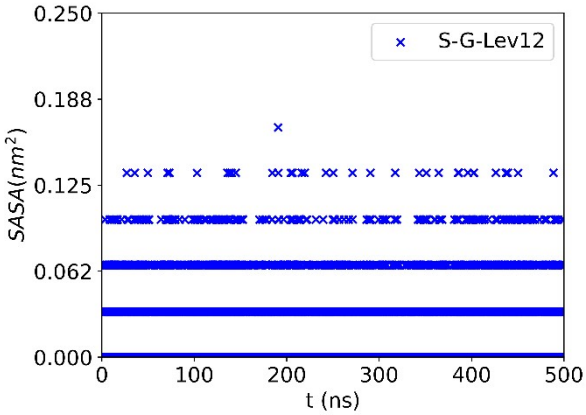


(c) G-S-Oxa11

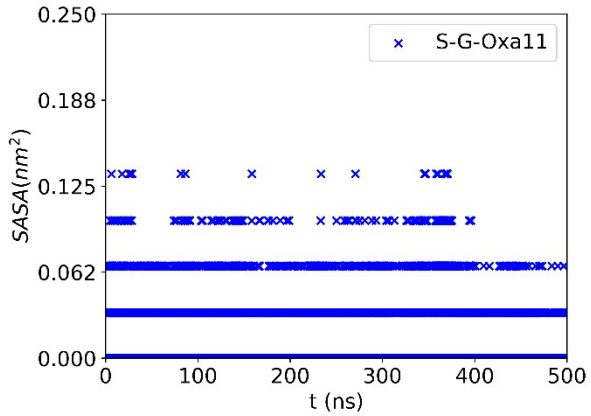
Figure S18. The SASA of G-S lignin dimer in the three DESs as a function of time.



(a) S-G-Lac12

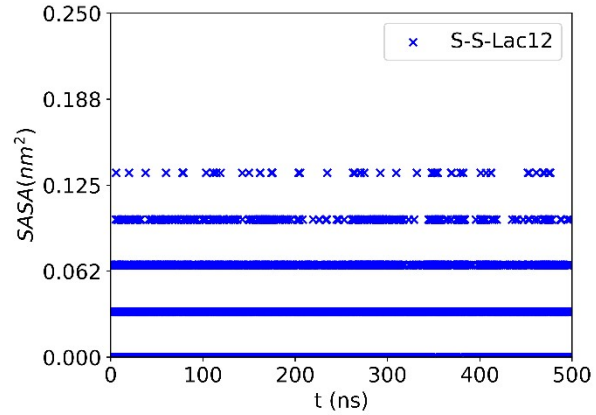


(b) S-G-Lev12

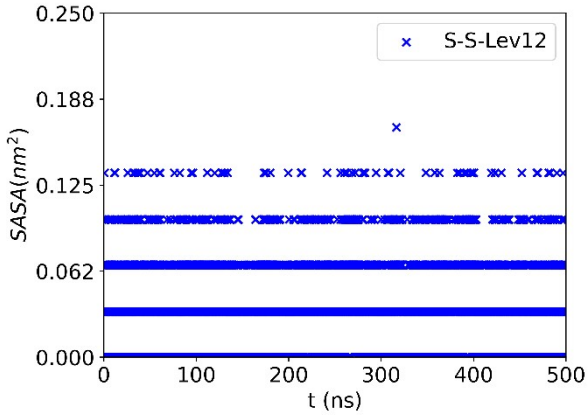


(c) S-G-Oxa11

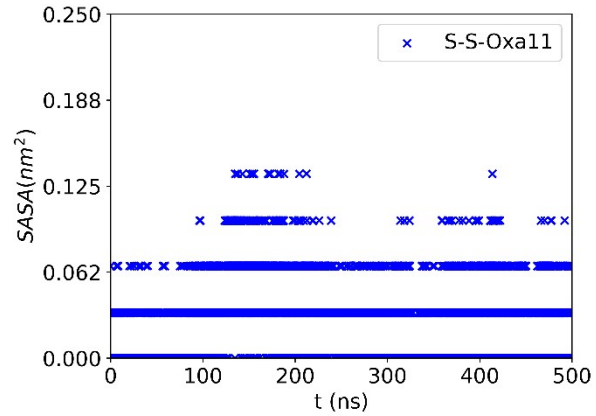
Figure S19. The SASA of S-G lignin dimer in the three DESs as a function of time.



(a) S-S-Lac12



(b) S-S-Lev12



(c) S-S-Oxa11

Figure S20. The SASA of S-S lignin dimer in the three DESs as a function of time.

Table S1. The percentage of a negatively charged solvation shell for the lignin dimers in the three DESs.

	% of negatively charged solvation shell		
	Lac12	Lev12	Oxa11
G-G	0.09%	0.63%	0
G-S	0.07%	0.28%	0
S-G	0.14%	0.45%	0
S-S	0.02%	0.39%	0.01%