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Ionic Liquids in Natural and Synthetic Polymers Upcycling

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Abstract

Limited biotic and abiotic technologies to process increasing deposits of synthetic polymers (such as polyolefins, polyesters, polyamides) and recalcitrant natural polymers (for instance, lignin predominantly from pulp and paper industries) remains a topic of discussion across the globe. In this regard, ionic liquids (ILs), also commonly described as "designer solvents" have attracted much attention for recycling and upcycling of these polymers. Based on the ease of structural modifications to tune the physicochemical properties of ILs, we designed and studied ILs to generate non-technical lignin from biomass with unique properties as discussed in this presentation. Furthermore, we also demonstrate the IL-catalyzed depolymerization of the generated lignin into bio-accessible intermediates to aid bioconversion. Lastly, we discuss an integrated one-pot IL-based hydrolysis of mixture of polyesters - polyethylene terephthalate (PET) and poly lactic acid (PLA) – followed by bioconversion of the depolymerized stream using *P. putida*.

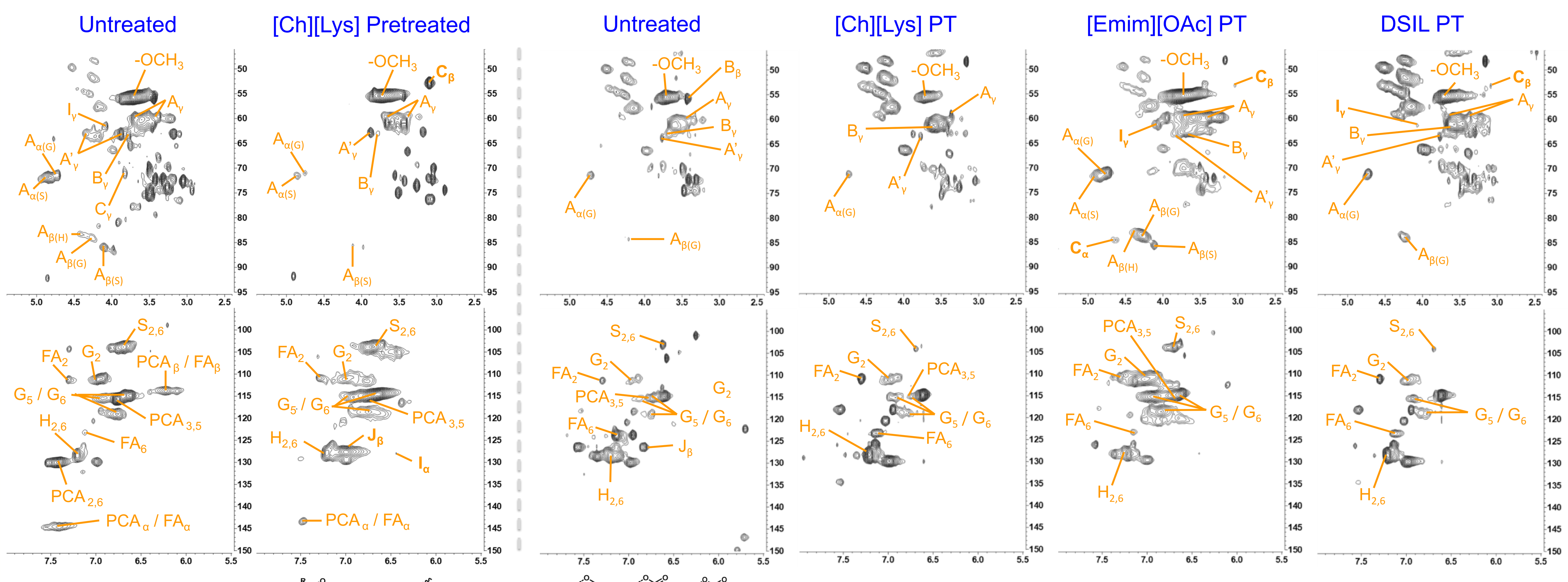
Objective

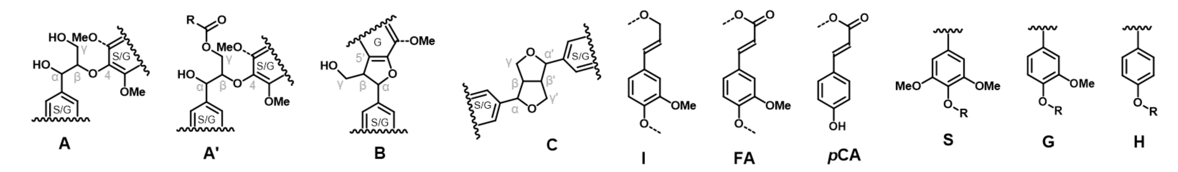
To develop and demonstrate integrated, feedstock agnostic, and efficient biotic and abiotic technologies using ionic liquids for

- replacing significant fraction of petroleum-derived fuels and chemicals with cost-competitive and renewable biofuels and bioproducts
- upcycling of abundant biomass (natural polymers) and plastic waste (synthetic polymers)
- utilization of whole biomass component including underutilized lignin
- high yields of intermediates compatible with downstream processing
- overall lower economic and environmental impact

Results

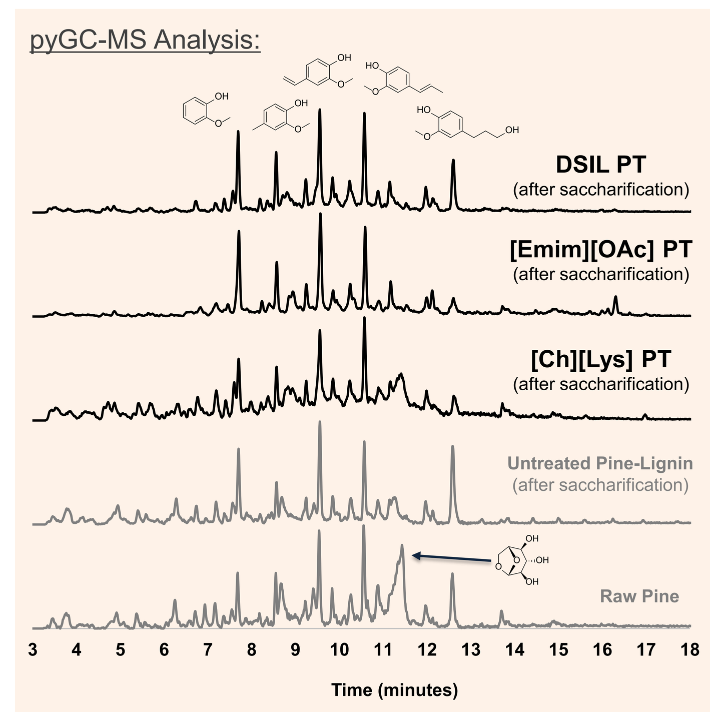
One-Pot Sorghum Lignin / *Pine Lignin Before and After Pretreatment*



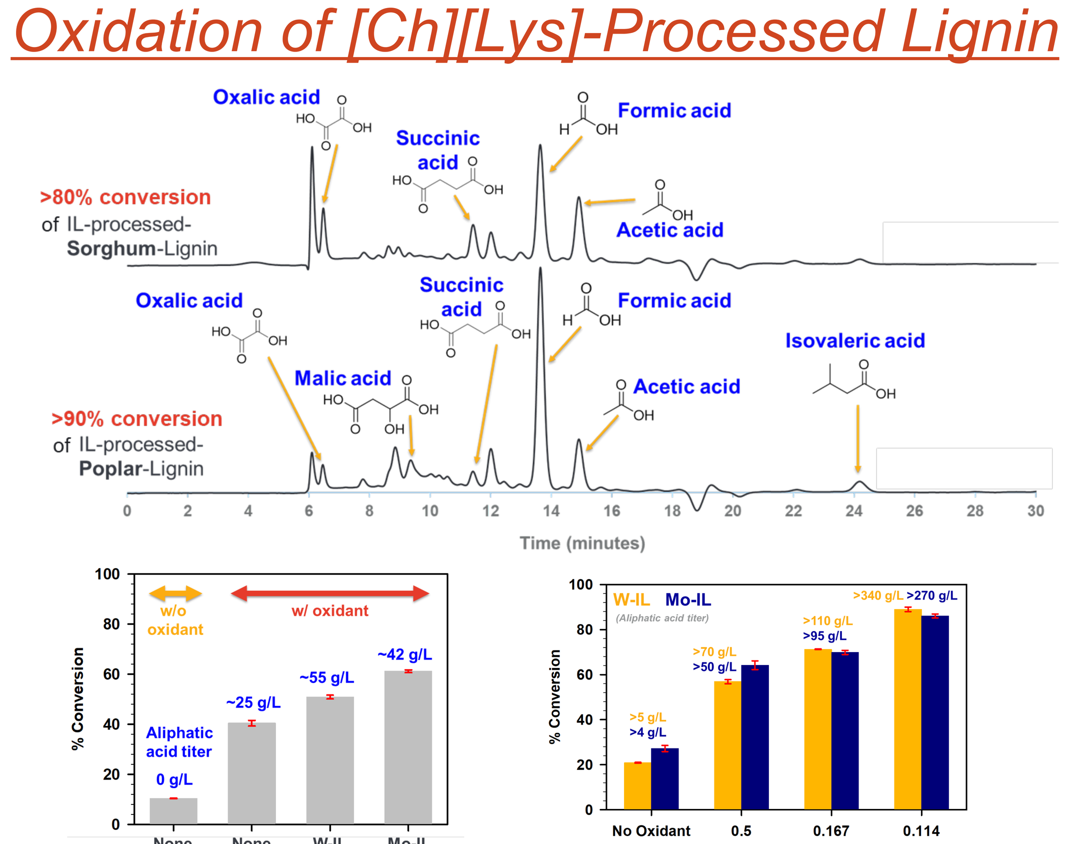


	%AIL[a]	M_w[b]	Units					Linkages		
			%S[c]	%G[c]	%H[c]	S/G	H/G	%C-C[c]	%C-O[c]	%OMe[c]
untreated	29.7	12817	6.0	64.0	30.0	0.09	0.47	7.3	16.5	76.3
[Ch][Lys] PT	42.3[d]	9626	1.3	66.3	32.4	0.02	0.49	14.4	17.4	68.3
[Emim][OAc] PT	75.9[d]	3873	7.4	82.4	10.2	0.09	0.12	11.9	18.2	69.9
DSIL PT	62.3[d]	6403	0.6	64.7	34.6	0.01	0.53	2.1	15.5	82.4

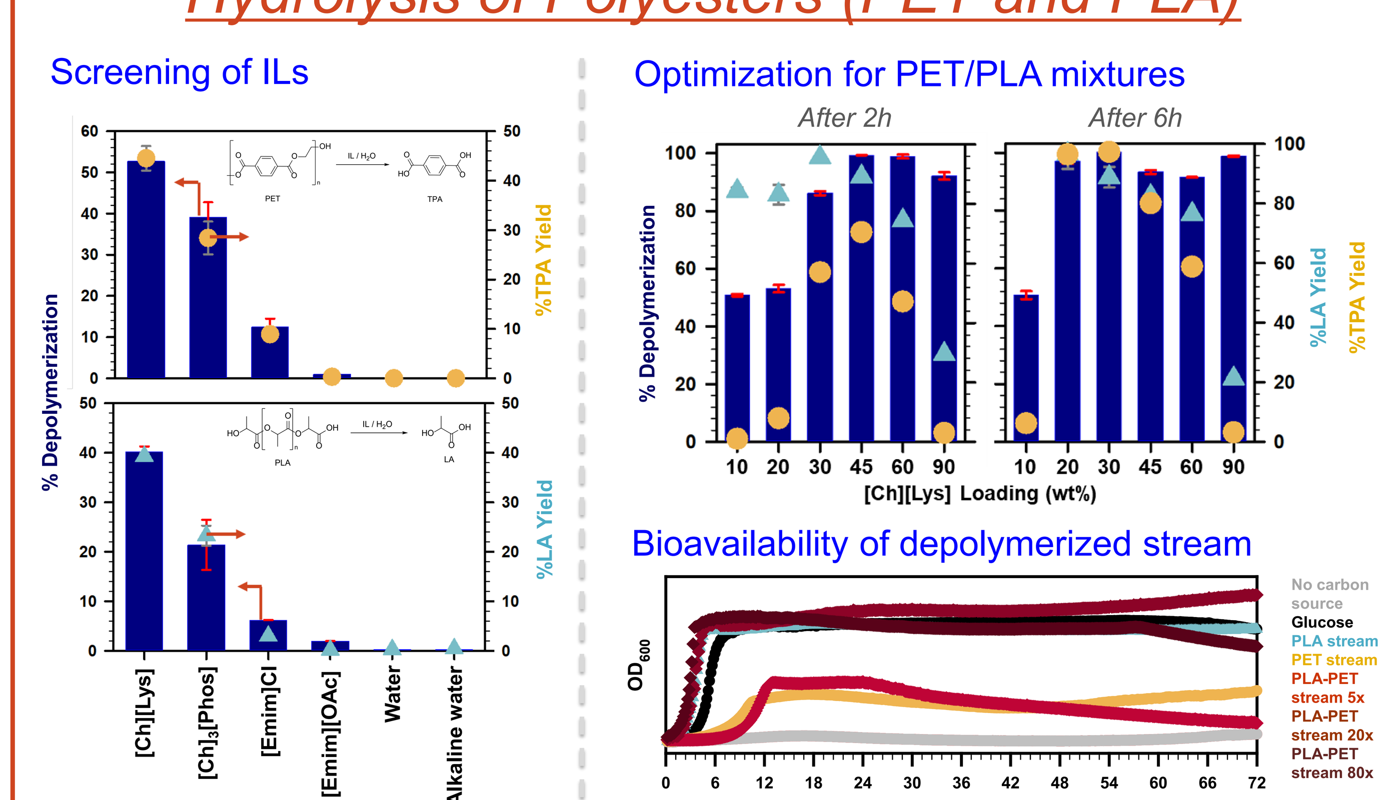
Pretreatment conditions: pine (20 wt%), IL (80 wt%), 140 °C, 3 h. [a]Acid-insoluble lignin. [b]Weighted average molecular weight based on GPC analysis. [c]Based on HSQC NMR. [d]Lignin content of the solid residue obtained after pretreatment and saccharification.



Oxidation of [Ch][Lys]-Processed Lignin



Hydrolysis of Polyesters (PET and PLA)



Conclusions

Ionic liquids serve as ideal solvent and/or catalyst system for the manipulation of both natural and synthetic polymers. Our study demonstrated that not only native-like lignin can be obtained after processing of holocellulosic fraction but also the generated lignin can be converted into bio-accessible organic acids by careful and precise design of ILs. Our results also emphasize that the use of biocompatible ILs can facilitate the bioconversion of plastic mixtures. This study demonstrates mere example of a potential IL-based approach for the upcycling of both natural and synthetic polymers. Attainment of overall sustainability and economic viability requires rigorous models based on structure-property / activity relationships (QSPR / QSAR) are required to understand the reaction pathways, catalytic sites, and environmental and economic impacts.

Acknowledgments

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