Autoxidation catalysis for C–C bond cleavage in lignin increases the theoretical aromatic monomer yield

Background

- Aromatic monomer yield from most lignin depolymerization strategies is solely from aryl-ether (C–O) bond cleavage, which limits aromatic monomer yields to ≤ 40 wt%.
- The added ability to conduct catalytic C–C bond cleavage in lignin would increase the theoretical monomer yield from lignin to near 100%.

Approach

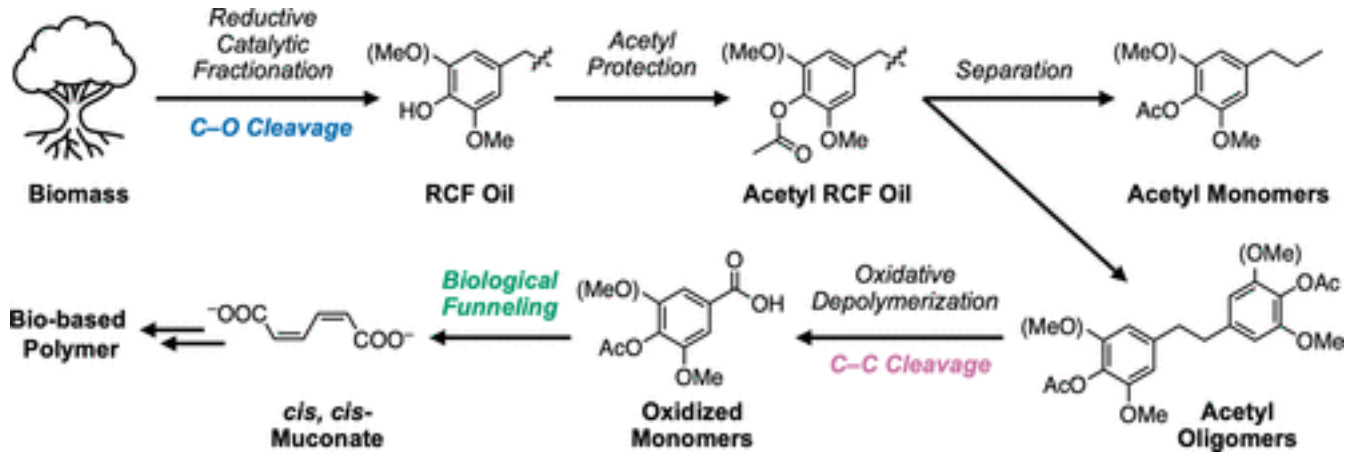
- We developed a catalytic autoxidation strategy to cleave C–C bonds in lignin-derived dimers and oligomers.

Results

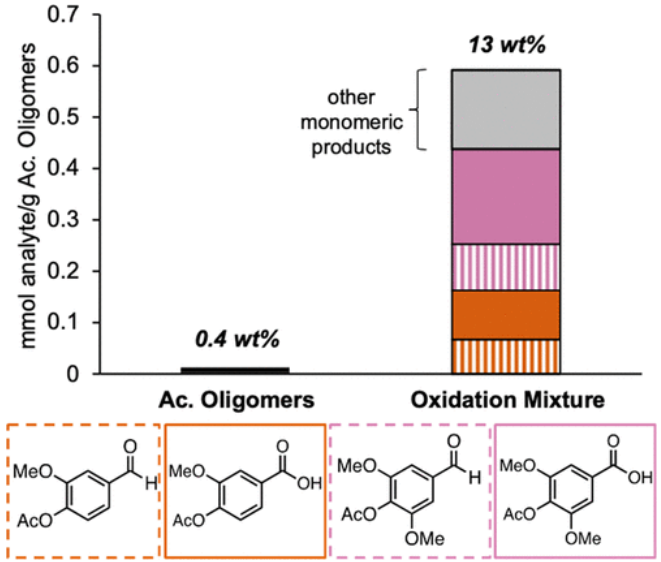
- The industrial autoxidation catalyst system, Co/Mn/Br in acetic acid with O_2 cleaves aliphatic C–C linkages in lignin model dimers with conversions of 87-99%.
- Acetylation is a technically viable strategy to overcome phenol inhibition in autoxidation catalysis.
- Lignin streams that contain only C–C bonded dimers and oligomers enable the production of bio-available aromatic monomers.
- Biological funneling experiments demonstrated that the oxidation products can be converted to muconic acid,

Significance

- This work demonstrates a C–C bond catalytic strategy to overcome the aromatic monomer yield ceiling set by C–O bond cleavage in lignin alone.



Overall lignin conversion approach featuring an oxidative C–C bond cleavage process to generate oxidized monomers that are suitable for biological funneling to a single product.



Monomer content in the starting acetyl oligomer material and resulting oxidation mixture (products quantified by UHPLC and LC-MS). Oxidation yields are an average of four runs. Wt % values expressed as the weight of total oxidation products/weight of acetyl oligomer substrate.

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