

Influence of Lignin Structural Architecture on the Performance of Lignin-Based Vitrimers

Background/Objective

- Vitrimers are novel polymers that combine the best durable features of thermoset plastics with potential recyclability. Although lignin can be utilized for vitrimer production, the lignin poses challenges in polymer thermomechanical performance.
- Here, we present a systematic investigation of how specific lignin structural features dictate thermomechanical performance, linking structure with performance and enabling predictable, tunable design of lignin-based polymers.

Approach

- Organosolv lignin was sequentially fractionated in ethyl acetate, ethanol, and acetone to obtain four lignin fractions with distinct structural characteristics.
- Lignin fractions modified through carboxylation were crosslinked with epoxidized soybean oil (ESO) to produce lignin-based transesterification vitrimers.
- Characterization of lignin fractions was performed, and thermomechanical performance and crosslinking density of vitrimer samples was performed.

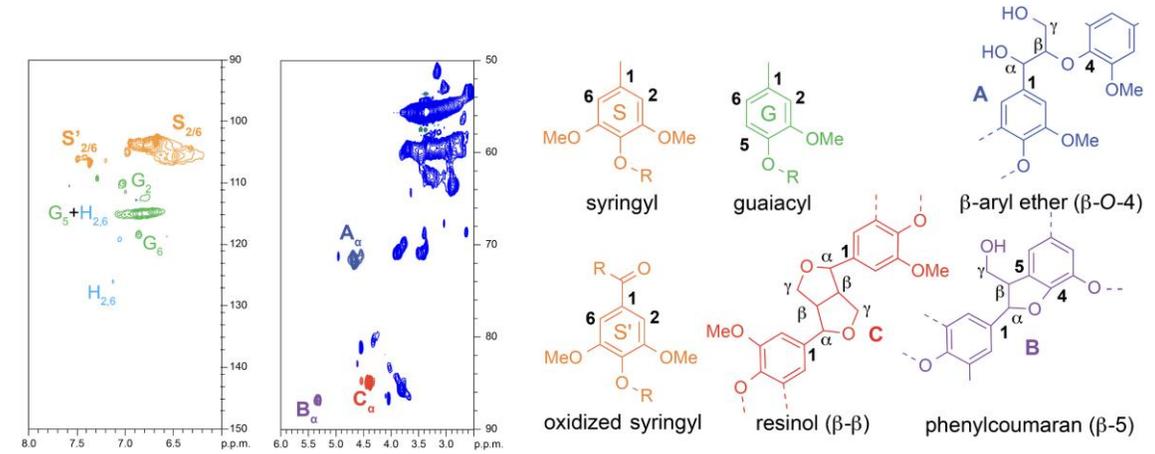
Results

- The hydroxyl content and the ratio of rigid to flexible interunit linkages in lignin showed a significant impact on the mechanical properties of lignin-based vitrimers.
- A positive relationship was observed between the thermal stability of the synthesized samples and molecular weight of lignin, while the energy demand for the bond exchange reaction decreased with increase in the OH content.

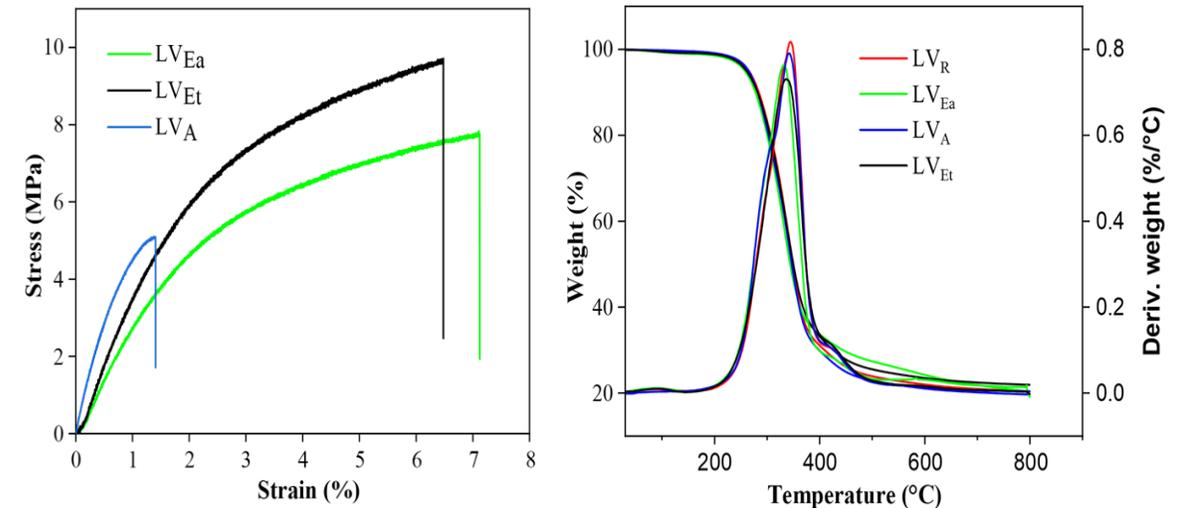
Significance/Impacts

- Lignin structure leads to better thermomechanical properties of lignin-based transesterification vitrimers. This will allow us to overcome lignin variability and design materials with tailored properties for targeted applications

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2-D heteronuclear single quantum coherence spectra highlighting the major structural units of lignin in aromatic (left) and oxygenated (right) regions.



Stress-strain curves (left) and thermal degradation (right) of synthesized vitrimers. LV_{Ea}, LV_{Et}, LV_A & LV_R represent lignin-based vitrimers derived from ethylacetate-soluble, ethanol-soluble, acetone-soluble, and residual lignin fractions, respectively.

